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Utilization of the forest industry sludges trends and effects on modern mills

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Report

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Utilization of the forest industry sludges – trends and effects on modern mills

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## Abstract

Katja Kuparinen, Satu Lipiäinen, Jussi Saari, Esa Vakkilainen Utilization of the forest industry sludges – trends and effects on modern mills Lappeenranta 2020 131 + 2 pages Report

The objective of this study is to discuss properties and treatment of the forest industry sludges. Primary, bio- and tertiary sludges are side streams formed in the pulp mill wastewater treatment. Their disposal and reuse is challenging due to sludge properties. Tightening legislation regarding waste treatment and disposal, and the growing need to increase efficiency in all the unit processes of a pulp mill act as driving forces to find environmental-friendly and energy-efficient ways for sludge treatment. Currently, sludges are often combusted in mill's boilers. In addition to energy production sludges can be used as soil improvement. This study compares the currently used methods for primary, bio- and tertiary sludge handling in kraft pulp mills and presents two potential new methods, namely, biogas production and hydrothermal carbonization (HTC). These processes are also presented in more detail by introducing mill example cases and via reference mill calculations. The study shows that HTC process could transform the biosludge treatment process from an energy consumer into an energy producer.

Keywords: biosludge, energy efficiency, hydrothermal carbonization, forest industry, energy consumption, energy production, best available technology

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# Appendix A: Biosludge properties

Appendix B: Sludge analysis results

# Nomenclature

#### **Roman letters**

Α	ash content	[%]
h	heating value	[MJ/kg]
'n	mass flow	[kg/s]
TCI	total capital investment	[€]
TS	total solids content	[%]

# Subscripts

evap	evaporation
eff	effective
ref	reference

## Abbreviations

AD	anaerobic digestion
ADt	air-dry tonne
AOX	adsorbable organic halides
BAT	best available technology
BDt	bone-dry tonne
BFB	bubbling fluidized bed (boiler)
BOD	biological oxygen demand
BREF	best available technology reference document
CaCO <sub>3</sub>	calcium carbonate
CaO	calcium oxide, lime
Ca(OH) <sub>2</sub>	calcium hydroxide, slaked lime
Cd	cadmium
CFB	circulating fluidized bed (boiler)
CH <sub>4</sub>	methane

CO	carbon monoxide
$CO_2$	carbon dioxide
COD	chemical oxygen demand
CTMP	chemi-thermomechanical pulp
ds	dry solids
EC	European Commission
ESP	electrostatic precipitator
EU	European Union
HCl	hydrogen chloride
HHV	higher heating value
HTC	hydrothermal carbonization
KCl	potassium chloride
LHV	lower heating value
MBBR	moving bed biofilm reactor
MP	medium pressure
NaCl	sodium chloride
NaOH	sodium hydroxide
Na <sub>2</sub> S	sodium sulphide
Na <sub>2</sub> CO <sub>3</sub>	sodium carbonate
NG	natural gas
NH <sub>3</sub>	ammonia
NO <sub>x</sub>	nitrogen oxides
NPE	non-process element
PAH	polycyclic aromatic hydrocarbon
PCB	polychlorinated biphenyl
PCDD	polychlorinated dibenzodioxins
SNCR	selective non-catalytic reduction
$SO_2$	sulphur dioxide
TCDD	tetrachlorodibenzodioxins
TSS	total suspended solids

# 1 Introduction

The forest industry is one of the world's largest industries. Broadly, it can be divided into the chemical and mechanical forest industry. The mechanical forest industry produces wooden products like sawn wood and wood-based panels. The chemical forest industry produces pulp, paper, and paperboard with several processes. Like many other industries, the forest industry generates significant amounts of byproducts and waste streams. Most of the byproducts and wastes, like black liquor, can be internally used in a profitable way. For some e.g. tall oil produced during the chemical pulping process, a biorefinery-based solution has been found. Tall oil can be hydrogenated to be used as transport fuel. Some byproduct streams, such as bark, fines, or sawdust, can be easily utilized for energy production at mill or outside the mill. At the moment, all end products cannot be reused or disposed of easily. Wastewater treatment in the pulp and paper industry produces biosludge, which is a good example of a challenging side stream that is neither a valuable raw material nor easy to dispose. Sludge disposal can account for over 50% of overall wastewater treatment costs and therefore, it is an important issue especially in chemical pulp mills (Meyer et al., 2018).

## 1.1 Chemical pulping

Chemical pulp is the major raw material of paper and paperboard grades. The production rate of chemical pulp is high globally. Sulphate (kraft) pulping is the most used pulping method and accounts for up to 80% of the global pulp production (Suhr et al., 2015). The used processes are complicated and generate a high amount of byproducts and waste streams.

The chemical pulping process is based on cooking of wood fibres using alkaline cooking liquor. The process uses softwood or hardwood as raw material and the final product can be bleached or unbleached. A wide variation of chemicals is used in the processes. Chemical pulping consists of several process stages, which are depicted in Figure 1.1. The process can be divided into the fibre line and the chemical recovery. The fibre line

consists of processes that are used for wood fibres treatment. The chemical recovery line can be divided further into the liquor cycle and the lime cycle.

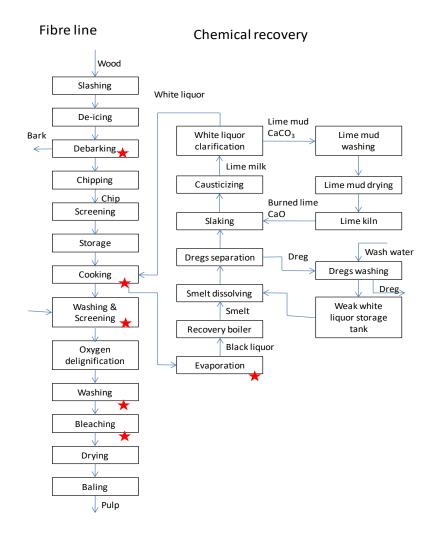


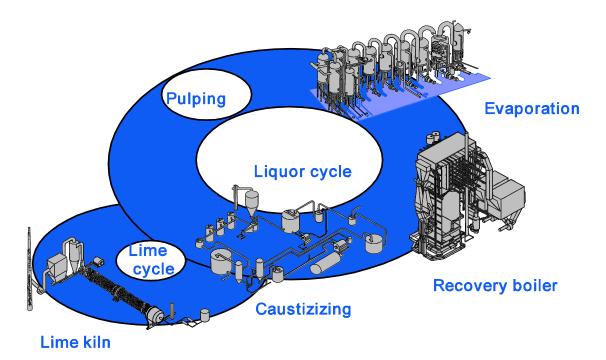
Figure 1.1. Processes in the chemical pulping. The red stars stand for processes that produce wastewater.

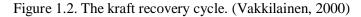
A kraft pulp mill consists of the fibre line and the chemical recovery cycle. The recovery cycle includes the liquor cycle and the lime cycle (Figure 1.2) and has the following functions:

- the recovery of the pulping chemicals
- the incineration of the dissolved organic material of black liquor in the recovery boiler to generate process steam and electrical power
- the recovery of organic byproducts (e.g. tall oil and lignin).

#### 1.1 Chemical pulping

The dry solids content of weak black liquor from pulp washing is 14–18%. Before combustion in the recovery boiler, the liquor is concentrated in the evaporation plant to the dry solids content of 65–80%, in modern mills, up to 80–85%.





Chemical pulping, mechanical pulping, and papermaking processes use a notable amount of water for dilution and washing, process steam, hot water, and cooling water. The red stars in the Figure 1.1 show the primary process stages that produce wastewater in kraft pulping. Cooling water is usually separated from the processes and therefore, does not need wastewater treatment. The major sources of wastewater in chemical pulping are bleaching, wood handling, foul condensates from evaporator and cooking, spillages, and washing losses. Most of the wastewater is produced in the bleaching stage. The amount of wastewater depends on the chosen processes. If wet debarking is used, the volume of wastewater is  $0.6-2 \text{ m}^3/\text{m}^3_{wood}$ . Dry debarking produces less wastewater, about  $0.1-0.5 \text{ m}^3/\text{m}^3_{wood}$ . Various condensates form  $8-10 \text{ m}^3/\text{ADt}$  of wastewater. Majority of condensates is recycled and reused in e.g. washing. Spillages occur in the digestion plant, the screening plant, during washing, in the evaporators and during causticizing. Modern

pulping collects and recycles spills even those formed during shutdown. Typically, 20– $25 \text{ m}^3/\text{ADt}$  of wastewater is produced in the new kraft pulp mills (Suhr et al., 2015). Chemical pulping is the major producer of biosludge in the forest industry.

The main purpose of evaporation is to increase the dry solids content of the black liquor. The other purpose is to separate methanol, turpentine and tall oil soap which are generated in the cooking phase. The evaporation stage consists of multiple-effect evaporators that are connected in series.

## **1.2** Sludge formation in the forest industry

Wastewater produced in the forest industry processes is purified by separating impurities from water using biological treatment. The reject is called sludge. Forest industry units produce a range of various types of sludge. Pulp and paper mills produce relatively high volume of primary sludge that consists mainly of short wood fibres. The aim is to reduce compounds that settle. Biosludge is produced in mills, which use active sludge wastewater purification process. In addition, deinking and chemical (tertiary) sludge can be produced. Properties of different sludges are presented in Table 1.1.

Parameter	Unit	Pulp mill sludge- mix	Primary sludge	Paper mill sludge- mix	Biosludge	Deinking sludge	Debarking sludge	Tertiary sludge
Moisture content	m-%	75-80	70		85	60	70	
Ash	m-%	13-21	25-60	12-20	16	30-60	2.5	
С	m-%	40-42	44	44-46	47	25-45	50	46-58
Η	m-%	4.5-5.0	6	5.5-6.0	5.2	4-5.5	б	6.4-7.2
S	m-%	0.4-1.3	0.1	0.05-0.1	1.2	0.1-0.3	0.02	0.5-2.4
N	m-%	1.3-2.9	0.4	0.5-0.7	1.6	0.1-0.3	0.8	0.3-2.3
0	m-%	25-29	25		30	22	34	
Cl	m-%	0.1-0.8		0-0.1	0.04-1.5	0.2-0.6		0.01-0.12
LHV	MJ/kg	14-18	2.3		17.4	8-13		16.8-22.3

Table 1.1. Main properties of various sludges (dry basis). (Lohiniva et al. 2001; Strömberg & Herstad Svärd, 2012)

Primary and secondary sludge are the major sludge types from modern wastewater handling. Primary sludge is formed in wastewater primary clarifier and it contains compounds that settle easily, such as fibres and other solids from pulping and papermaking processes. Secondary sludge (biosludge) is produced in secondary clarifiers in mills with biological wastewater treatment plant. Figure 1.3 shows the simplified forming process of primary and secondary sludge in a wastewater treatment plant.

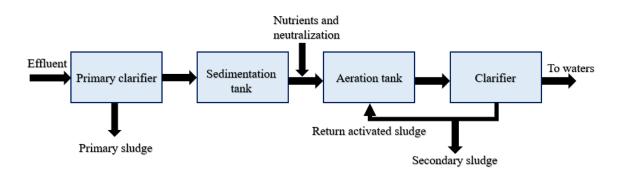


Figure 1.3. Separation of primary and secondary sludge in a wastewater treatment plant. (Modified from Liimatainen, 2000)

Tertiary sludge, also called chemical sludge, originates from additional chemical treatment of wastewater (Hynninen and Dahl, 2008). The purpose of tertiary treatment is to remove contaminants that the preceding steps did not sufficiently remove, such as colour or metals (Suhr et al., 2015). Tertiary treatment has typically low additional environmental benefits to the state of the recipient waters (Sandström et al. 2016). Sometimes due to sizing of preceding treatments, an additional stage is required.

Sludge has a high moisture content, which can make its disposal challenging. Moisture is relatively easy to remove from the fibre-rich primary sludge by e.g. pressing. Therefore, primary sludge can be combusted in the boilers in forest industry mills. Biosludge disposal is more complicated due to its properties that make dewatering difficult.

Forest industry units produce a notable amount of sludge during normal operation. In the year 2014, 537 000 tons of sludge was produced in the Finnish mills (Alakangas et al.,

2016). In 2017, about 3 200 dry tons of sludge ended up in landfills (Metsäteollisuus, 2019).

The composition of forest industry sludge differs from the composition of municipal wastewater sludge due to different types of processes. Secondary sludge from pulp and paper mills has some similar characteristics with municipal activated sludge. Nutrient values, such as nitrogen, phosphorus, and potassium, vary depending on the mill process. Nitrogen and phosphorus levels are typically lower in pulp and paper mill sludges than in municipal sludge (Lohiniva et al., 2001), while calcium and magnesium levels tend to be higher (Bajpai, 2015). Heavy metal contents are often smaller in pulp and paper mill sludges than in municipal sludge, and unlike municipal sludge, they usually do not include intestinal bacteria (Lohiniva et al., 2001). Mixing with sewage sludge has been seen as a low-cost option to increase the nutrient status of pulp and paper mill sludges in anaerobic digestion trials; though, anaerobic digestion is not usually applied for pulp and paper mills sludges due to long residence times (Bajpai, 2015).

Sludge disposal has become more and more difficult due to higher wastewater purity requirements, higher share of biosludge, and tightening legislation that limits the possible disposal methods. For example, in Finland landfill disposal of organic wastes is prohibited; waste materials should be recycled, reused as raw material, or disposed of as an energy source. As the amount of biosludge has increased, as well as its share of the total sludge, the handling of mixed sludge has become more challenging. Therefore, reasonable disposal methods for especially biosludge are needed.

Another problem, especially in the southern hemisphere is the fact that sludges during storage tend to decompose and form during decomposition unpleasant odour. Restricting this odour has also been a subject of environmental permits given. Using biosludge as soil amendment has several restrictions for it to be effective (Feagley et al., 1994; Norrie & Gosselin, 1996; Mahmood & Elliot, 1996)

This report looks at modern sludge disposal methods and therefore possible use as soil improvement or lining material is not discussed.

## 2 Primary sludge

Pulp mill wastewater contains small fibres and suspended larger solid particles. The primary sludge is produced in the primary clarifier of a wastewater purification plant where the solid matter is settled in a clarifier thus generating primary sludge. Typically, the residue is removed from the bottom and prepressed.

Unlike many other pulp mill waste streams, the amount of primary sludge can be reduced by minimizing the amount of fibre reject entering the wastewater system (Suhr et al., 2015). Especially pumping all, even small streams, from the processing area drains to evaporator instead to wastewater treatment has reduced primary sludge. Primary sludge production in a kraft pulp mill can be at 2–13 kgds/ADt.

## 2.1 **Primary sludge properties**

Primary sludge consists of mainly of fibres and fibre fragments and it often includes also bark, paper additives, fillers, and grit (Hynninen & Dahl, 2008; Suhr et al., 2015). Water can be in free, capillary, or intercellular forms. Sludge properties are strongly dependent on the mill process and products. Table 2.1 represents example compositions of primary sludge based on references.

	Faubert et al. (2010)	Lohiniva et al. (2001)	Strömberg & Herstad Svärd (2012)	Zhang et al. (2010)
С		44	47.5	40.3
Н		6	6.6	5.17
Ν	0.045-0.28	0.4	0.44	3.28
S		0.1	0.24	0.252
Na				0.241
Cl			0.02	
Κ	0.02-0.09			0.340
Ash	10–15	25-60	14.0	21.5

Table 2.1. Example compositions of primary sludge dry matter (m-%) based on references

Typically, ash content varies between 3% to 60%, the lowest content being in pulp mill primary sludge and the highest in fine paper mills (Lohiniva et al., 2001). After mechanical dewatering, primary sludge can contain 35–40% dry solids and its lower heating value (LHV) is at 4–6 MJ/kg.

## 2.2 Primary sludge handling

The main purpose of primary sludge treatment processes is dewatering to enable subsequent processing or use. Primary sludge is typically combusted in mill boilers after dewatering. It is well suitable for combustion due to its high content of fibres. Sludge treatment can include thickening, conditioning, dewatering, and drying (Bajpai, 2015).

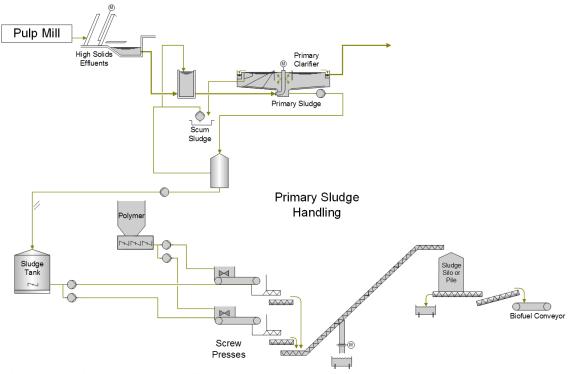


Figure 2.1. Primary sludge handling.

Primary sludge is easier to handle and dewater than especially secondary sludges. In many mills other sludges are mixed with primary sludge to minimize the challenges caused by their poor dewatering properties. Separate handling of each different type of sludge would make it possible to choose the best processes based on sludge properties and choice of further utilization, which might enable more utilization options and additional revenue. Primary sludge can be e.g. used as additional material in the brick industry (Suhr et al., 2015).

Typically, sludge is thickened and dewatered mechanically. Conditioning can be done but unlike with mixed or biosludge, it is not always required for primary sludge. Typically, thermal or reactant conditioning treatments are not used (Bajpai, 2015). Reactant treatment includes chemicals that are used to form flocs to improve dewatering.

Most of free water in primary sludge can be removed by gravity settling; solids sink to the bottom of the tank. Capillary water is removed mechanically. Belt press, filter press, screw press, vacuum filter, or centrifuge are the most often used mechanical dewatering devices. Mechanical dewatering equipment is introduced in more detail in Section 3.2.2 that discusses mechanical dewatering of biosludge. Sometimes different presses can be used sequentially.

Sometimes sludge is thermally dried after mechanical dewatering. Drying decreases volume and increases the heating value of sludge; wet sludge has a negative heating value. Excess heat use for drying can reduce costs and improve process efficiency. Thermally dried primary sludge is easy to handle and store (Lohiniva et al., 2001). Section 3.2.3 discusses thermal drying methods.

# 3 Biosludge

The secondary sludge, also called biosludge, is produced in the secondary clarifier of an aerobic wastewater treatment plant. Wastewater treatment plants in the forest industry mills utilize micro-organisms to purify the water. These organisms feed on the organic materials in the wastewater and transform wastewater impurities into carbon dioxide, nitrogen, and microbial mass (Liimatainen, 2000). Biosludge is produced when excess, spent microbial mass settles to the bottom of the pond. Biosludge consists of the micro-organisms, such as organic dead bacteria, unsettled fibres, and undigested organics (Hovey, 2016). Organic material is the main component of biosludge, about 65–70% of the mass (Alakangas et al., 2016). The cells of the micro-organisms include water and consequently, the structure of biosludge complicates the dewatering significantly. Only very low dry solid contents are therefore reached. This means that biosludge is a challenging product to reuse, dispose of, or transport. Typical amounts of biosludge produced in a stand-alone pulp mill and an integrated pulp and paper mill are presented in Table 3.1. Often cited 15 kg/ADt for chemical pulp might be excessive in modern plants where bleaching is optimized, and spills are diligently collected and recycled.

Product	Biosludge [kg/tproduct]
Paper and paperboard	1–5
Pulp	10–28
Mechanical pulp	8–15

Table 3.1. Biosludge production (Lehtinen, 2001)

## 3.1 **Biosludge properties**

Biosludge consists mainly of dead organisms, but it also contains some inorganic materials. Typically, 15–20% of biosludge is inorganic material, usually reported as ash (Liimatainen, 2000). Example compositions of biosludge are collected in Table 3.2. The composition varies between mills and processes. Also compounds in water, such as some metals, concentrate in biosludge. About half of dry biosludge is burnable components (carbon and hydrogen). In addition to burnable components and ash, biosludge contains a large variation of elements, for example alkali metals, such as sodium. Some of the

components are valuable when reuse is considered, such as nitrogen, potassium, and phosphorus that are important in fertilizers. On the other hand, components like chlorine can cause problems for example in handling devices. The table below presents only the most significant components. More detailed biosludge compositions from Scandinavian mills are presented in Appendixes A and B.

	Vakkilainen & Pekkanen (2002)	Liimatainen (2000)	Harila & Kivilinna (1999)	Lohiniva et al. (2001)	Strömberg & Herstad Svärd (2012)	Zhang et al. (2010)
С	50.4	45-47	47	47	53.0	41.2
Н	6.0	5.4-6.5	6-6.5	5.2	7.0	4.51
Ν	0.7	1.5-4.7	3.4-4.3	1.6	4.4	4.18
S	3.7	1.2-3.8	1.2-2.2	1.2	1.8	2.33
Na	9.7	0.4-1.6	0.43-0.50	-		
Cl	2.5	0.1-0.7	0.0001- 0.0002	0.04-1.5	0.1	
Κ	0.16	0.1-0.3	0.16-0.32	-		
Ash	-	10-20	10-15	16	2-60	24.4

Table 3.2. Example compositions of biosludge (m-%) according to references.

The heating value of dry biosludge varies depending on the sludge composition. According to Vakkilainen & Pekkanen (2002), the higher heating value of biosludge can be at 20 MJ/kg. Although the heating value of dry and ash-free biosludge is relatively high, due to high moisture and ash content, the heating value as fired is low, at about 0.5 MJ/kg (Alakangas et al., 2016). The heating values for biosludges with different dry solid and ash contents can be calculated using the following equation (Hagelqvist, 2009):

$$h_{net} = h_{eff} \cdot \left(1 - \frac{A}{100}\right) \cdot \frac{TS}{100} - h_{evap} \cdot \left(1 - \frac{TS}{100}\right)$$
(1)

Where

$h_{net}$	Heating value	[MJ/kg]
$h_{\it eff}$	Effective heating value of dry, ash-free fuel	[MJ/kg]
Α	Ash content	[%]
TS	Total solids content	[%]
$h_{evap}$	Evaporation heat of water	[MJ/kg]

The heating value is strongly related to the moisture content (Figure 3.1). Therefore, combustion properties of biosludge are often poor due to low dry solids content. Typically, slightly over 10% dry solids content is reached with a reasonable amount of effort and costs. The dry solids content of pre-treated but not mechanically dewatered biosludge is usually below 5%. Figure 3.1 shows that the lower heating value is then negative.

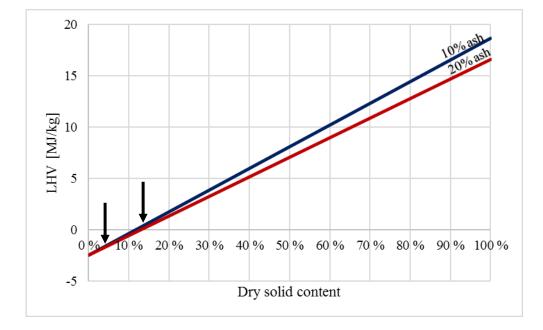


Figure 3.1. The lower heating value of biosludge as a function of dry solid and ash content. The arrows show typical dry solid contents before and after dewatering.

High moisture content is the most significant feature of biosludge. In wastewater purification plants, the dry solid content after settling is 5–15 g/l (Vakkilainen & Pekkanen, 2002), which is about 0.5–1.5%. Water is bound into biosludge particles in different ways, as Figure 3.2 shows. About 70–75% of water is free water that is not bound in the solid material and can therefore be relatively easily separated using gravity filtration (Forsell-Tattari, 2013; Hovey, 2016). Interstitial water is stored within the sludge flocs by capillary forces and can be removed only by breaking the cell walls. Surface water is bound to the surface of the sludge particles by hydrogen bonds.

Intracellular and chemically bound water is inside the cells, which makes dewatering challenging. Both surface and intracellular water can be removed only by destroying the cells thermally (Forsell-Tattari, 2013).

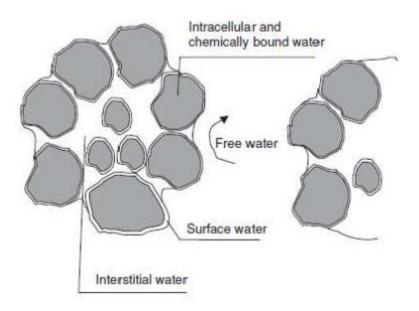


Figure 3.2. Distribution of water in biosludge particle. (Hovey, 2016)

Due to biosludge structure and its poor dewatering properties, only low dry solid contents can be reached by pressing. Pure biosludge can theoretically be dewatered to dry solid content of 10–15% (Vakkilainen, 2007). In many cases, supporting material such as primary sludge, is used to improve the handling properties of biosludge. With supporting material, higher average dry solids contents can be reached, but the biosludge fraction dry solids is still low. Increasing the amount of biosludge acts as a driving force for handling pure biosludge without mixing it to primary sludge.

Biosludge is typically odorous, which further complicates its treatment and disposal. Odours are harmful to both health and air quality and therefore, biosludge handling requires odour control. For example, stabilization processes reduce odours.

Detailed analyses on the composition of biosludge in Scandinavian mills (Appendixes A and B) show that biosludge contains nutrients such as N, K, Na, S, Ca, Mg, and P, which

indicates that sludges could be of use as fertilizers or in soil management after a suitable treatment. Sludges also contain low amounts of heavy metals, such as Zn and Pb. The permissible content of heavy metals in fertilizers and soil management is limited (Ojanen, 2001), but the analysis results show that their concentration in biosludge is notably under the set limits in most cases, as can be seen later in Table 7.2. A more significant issue regarding the reuse of sludges in agriculture is the high moisture content.

## 3.2 **Biosludge handling**

Biosludge processes aim at removing water from biosludge to make the sludge more usable for reuse, energy production or to other forms of disposal. Some of the processes attempt to mitigate pathogens, odours, health hazards, and environmental hazards, as well as to stabilize the sludge. The most significant dewatering is done with mechanical devices. Before the mechanical dewatering, pre-treatment processes like thickening, stabilization, or conditioning are utilized. Figure 3.3 presents possible biosludge treatment processes.

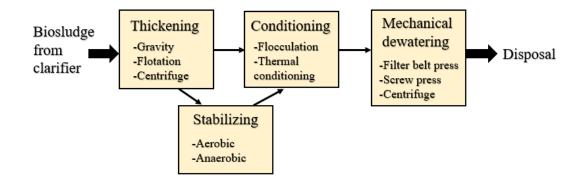


Figure 3.3. Biosludge treatment processes.

#### 3.2.1 **Pre-treatment processes**

Thickening is typically executed by gravity. Biosludge can be thickened either alone or with primary sludge. If biosludge is thickened alone, the dry solids content increases from 0.5–1.5% to 3–4% during this process stage (Liimatainen, 2000). In addition to the gravity method, also flotation and centrifuges can be utilized (Jaakkola, 1993).

The aim of stabilization is to improve the hygiene of sludge and reduce odours. Sludge can be stabilized using aerobic or anaerobic processes, composting, calcium stabilizing or heat treatment. (Lohiniva et al., 2001).

Before mechanical dewatering, there is a conditioning stage to sludge's handling properties during dewatering (Jaakkola, 1993). Flocculation is a widely used conditioning process, which raises the particle size of sludge using polymers. Higher particle size enhances dewatering properties. Usually flocculation is executed chemically, which means that chemicals are added to sludge during the process (Liimatainen, 2000). Typically, about 1–5 kg/tds conditioning chemicals are needed (Lohiniva et al., 2001). Thermal conditioning can also improve sludge's dewatering properties (Jaakkola, 1993).

#### 3.2.2 Mechanical dewatering

The aim of mechanical dewatering process is to increase dry solid content of sludge to as high as possible. High dry solid content increases heat available in combustion and reduces volume of sludge and water vapor in flue gases. (Liimatainen, 2000)

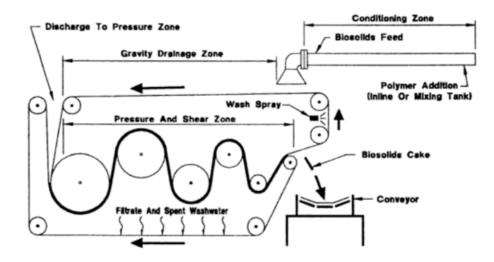


Figure 3.4. Belt filter press. (Edler, 2014)

Belt filter press (Figure 3.4) is the most used dewatering device in the Finnish forest industry (Ojanen, 2001). It is more suitable for primary sludge or mixed sludge than for

biosludge. Depending on the thickness of the belt, biosludge passes the device while most of the water or drains through the belt. Dry solid contents of 10–20% are reached with belt filter presses when biosludge is dewatered (Liimatainen, 2000). Some supportive material, e.g. peat, must be added to enable an effective pressing process (Lohiniva et al., 2001).

Figure 3.5 presents the achieved sludge dry solid content by belt filter press as a function of biosludge content. The figure shows that with filter press dry solid content of above 35% can be reached with pure primary sludge. Adding biosludge to primary sludge decreases the final dry solid content almost linearly, and the results of dewatering for pure biosludge are poor compared with pure primary sludge. Typically in practical mill operations, the final biosludge dry solids content is approximately 10 % or less. Often additive usage is required.

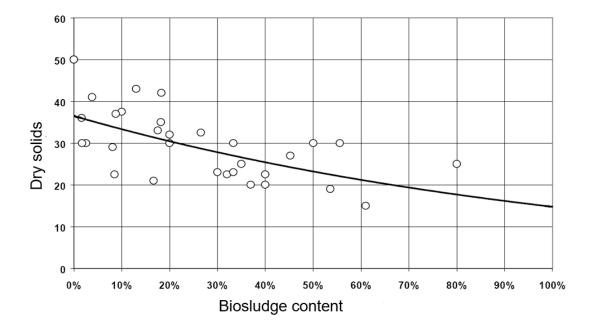


Figure 3.5. Dry solid content reached with belt filter press as a function of biosludge content. (Dahlbom. & Wadsborn. 2005)

Screw press is a mechanical dewatering device that consists of a cylinder and a screw. The cylinder is perforated, and when the screw presses sludge against the walls of the cylinder, the water exits through the holes (Forssell-Tattari, 2016). The device is typically used for primary and mixed sludge, but it can also be used for biosludge. Water inside the cells of biosludge reduce the process efficiency, but if steam is used to heat up the device, dry solid content of 10–15% can be reached. (Liimatainen, 2000). Figure 3.6 shows the principle of a screw press.

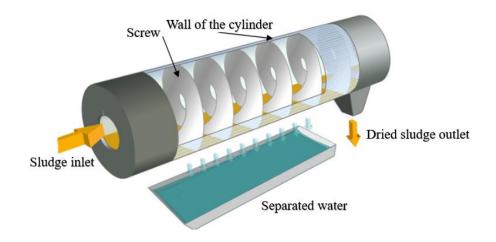


Figure 3.6. Screw press. (Modified from Forssell-Tattari, 2016)

Centrifuge is one of the most suitable devices when pure biosludge must be dewatered. The most common application is a decanter centrifuge (Figure 3.7). The process is based on high speed rotation. Due to rotation, heavier sludge particles are flung close to wall while water is more freely moved. Water is removed from one outlet and the dried sludge from another. A centrifuge typically reaches good dewatering results, has a high capacity, and requires relatively little space. Its disadvantage is high energy consumption (Motiva, 2018).

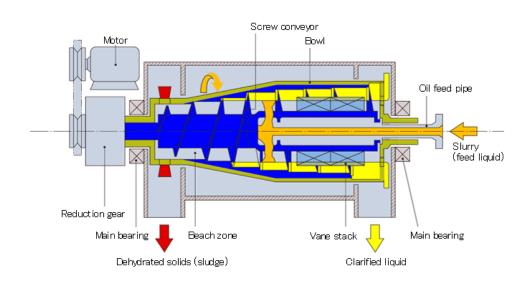


Figure 3.7. Decanter centrifuge. (Mitsubishi, 2019)

### 3.2.3 Thermal drying

Thermal energy can be used to evaporate moisture in sludge. The problem is that heat is often costly, and the benefits may be less valuable than the used energy. However, drying is obligatory with some disposal methods. For example, efficient incineration usually requires that dry solid content of biosludge must be at least 30–35% (Hovey, 2016). Waste heat utilization can make the drying process more feasible.

The thermal drying can be executed either directly or indirectly. In direct processes, sludge is in contact with the heating device and heat is transferred by convection. In indirect methods, the drying occurs through heat transfer surface by conduction or radiation. Rotary disc and rotary tray are examples of indirect drying methods. Direct methods include e.g. rotary drum, flash, and moving belt dryers (Hovey, 2016).

Paddle dryers are sometimes used for thermal drying of sludge. They are indirect dryers whose benefit in sludge drying is the use of mechanical agitation that helps to overcome stickiness (Mujumbar, 2015). Paddle dryers are relatively compact and energy efficient.

Thermal drying is not usually profitable for biosludge due to low dry solid content before the dryer, high steam consumption, and possible grabbing risk (Liimatainen, 2000). In addition, behaviour of biosludge during drying is challenging. When dry solid content of biosludge increases, the sludge becomes sticky. The stickiness causes agglomerating and adhering in the dryer, which decreases the process efficiency. Heat transfer coefficient can be even 60% lower with sticky materials compared with non-sticky materials. Besides the stickiness problems, biosludge emits odours during drying and causes corrosion risk on drying devices because the corrosive compounds can concentrate (Hovey, 2016).

Mäkelä et al. (2017) dried pure biosludge in pilot-scale experiments. The pilot set-up is presented in Figure 3.8. Dry solid content of biosludge feed was 9%, and it increased to 19–68% during drying with secondary heat. The reasons for variation in dry solid content were varying temperatures and sludge feeding rates. Energy consumption of drying was 0.6–1.7 kWh/kg H<sub>2</sub>O. The experiment indicates that biosludge can be dried using waste heat at pulp and paper mills. With waste heat temperature of 70 °C, sludge feeding capacity of 170 kg/h can be reached. After drying, biosludge can be used efficiently to replace fossil fuels.

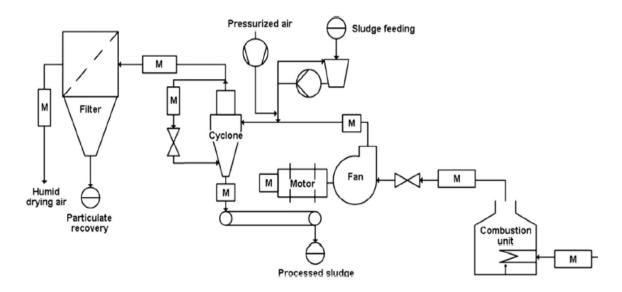


Figure 3.8. Pilot set-up of biosludge drying system. (Mäkelä et al., 2017)

## 4 Tertiary sludge

Tertiary sludge processes aim according to (Suhr et al., 2015) at removing contaminants that preceding treatment steps (e.g. biological treatment) could not remove or could not sufficiently remove, like color, metals, total suspended solids (TSS), chemical oxygen demand (COD), nitrogen or phosphorus. The main focus has been reducing toxicity and color from the wastewater after secondary wastewater treatment (Cabera, 2017). Main problem with tertiary sludge is when chemicals are used, the sludge contains problematic compounds. Tertiary treatment using activated carbon can also be used to remove metals from wastewaters (Alatalo et al., 2013).

Tertiary treatment is used in some mills and usually the amount of tertiary sludge is smaller than the amounts of primary and secondary sludges. A tertiary treatment process can be for example chemical precipitation followed by filtration or clarification to remove the formed flocs. Typically used chemicals are aluminium salts, ferric chloride, ferric sulphate, ferrous sulphate, or lime. Tertiary sludge formation, properties and handling is a little studied subject.

### 4.1 Tertiary sludge properties

Tertiary sludge consists mainly of flocs that are formed by the used coagulants in the treatment (Hynninen and Dahl, 2008). The composition, e.g. the amount of organic or inorganic material depends on the dosage and the chosen chemicals (Suhr et al., 2015). Chemical treatment can be used in several stages and also together with biological treatment, and therefore the compositions of the sludge varies greatly (Strömberg & Herstad Svärd, 2012). Table 4.1 presents analysis results on tertiary sludge.

		Strömberg & Herstad Svärd, 2012
С	m-%	45.8-58.0
Н	m-%	6.4-7.2
Ν	m-%	0.31-2.3
S	m-%	0.51-2.4
Cl	m-%	0.01-0.12
Ash	m-%	7.3-22.2
LHV	MJ/kg	16.8-22.3

Table 4.1. Analysis results on tertiary sludge (dry matter).

## 4.2 Tertiary sludge handling

Tertiary sludge can be even more difficult to handle than biosludge (Suhr et al., 2015). It can include residues of iron and aluminium in small amounts. Its composition is often fragile (Lohiniva et al., 2001), and similarly as biosludge, tertiary sludge has poor dewatering properties compared with primary sludge. Due to wide variation of properties, the behaviour of tertiary sludge varies greatly, which affects handling (Strömberg & Herstad Svärd, 2012).

Tertiary sludge cannot be combusted without auxiliary fuel due to high content of inorganics and water. It can somewhat be dewatered by use of centrifuge. If only synthetic organic polyelectrolytes are used for flocculation, sludge can be combusted; otherwise it is often landfilled (Suhr et al., 2015). However, it seems that combustion of tertiary sludge does not notably increase the risk of e.g. corrosive depositions in boilers (Strömberg & Herstad Svärd, 2012).

Because tertiary treatment is still rare, the processes used still vary a lot. Main processes considered for tertiary treatment in pulp and paper industry are membrane filtration, chemical precipitation, adsorption (activated carbon, bentonite, biofiltration, ion exchange resins) and ozonation (Hubbe et al. 2016).

### 4.2.1 Chemical precipitation

Using chemical to flocculate dissolved and suspended matter is long known to be applicable. Adding aluminium salts  $(Al_2(SO_4)_3, Al_n(OH)_mCl_{3n-m})$ , iron compounds (ferric chloride (FeCl<sub>3</sub>), ferric sulphate (Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>), ferrous sulphate (FeSO<sub>4</sub>) ) or lime (Ca(OH)<sub>2</sub>) generates solid residue that can be separated by clarification.

Only a few pulp mills in the world have installed tertiary treatment that uses chemical precipitation (Krogerus, 2016). In Nordics, Stora Enso Varkaus, Holmen Iggesund and Metsä Fibre Äänekoski use tertiary treatment where chemicals are added at tertiary stage. In addition, a few examples are found in Chile, Brazil and China. Figure 4.1 shows an example configuration of a tertiary sludge system using chemical precipitation.

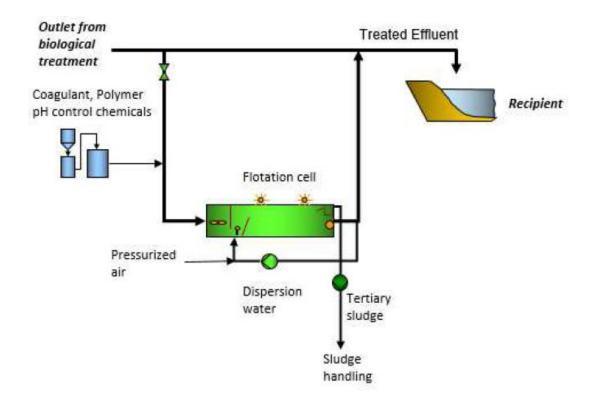


Figure 4.1. Tertiary sludge system using chemical precipitation. (Krogerus, 2016)

#### 4.2.2 Adsorption

One can also utilize materials with high surface are and affinity (activated carbon, ion exchange resin, processed biomaterials etc.) to adsorb contaminants from wastewater. Activated carbon has widely been used as an adsorbent in wastewater water treatment because it is highly efficient in removing organic compounds, metals and other inorganic pollutants (Alatalo et al., 2013). Activated carbon materials are not cheap as these carbon-based materials have increased porosity, large surface area, variable surface chemistry, and high degree of surface, which all help in capturing unwanted compounds.

Biosludge can be modified to work like activated carbon. The main advantage would be of removing unwanted heavy metals and other harmful but low content residues by replacing the expensive activated carbon by low cost adsorbent (Babel & Kurniawan, 2003). An additional advantage would be the possibility of disposing the spent biosludge the same way as before. i.e. biosludge would be sidetracked to be used to capture metals and then utilized as before. The disadvantage would be lower adsorption capacity. Usage of biosludge as absorbents has not been tried at full scale, but laboratory trials have been conducted where high capabilities to remove e.g. P and Cr were observed.

### 4.2.3 Additional filtration

In older mills with increasing production, tertiary units have been added as an alternative to increasing the hydraulic capacity of primary and secondary treatment. For the same purpose, also biofiltration has been used (Suhr et al., 2015). Figure 4.2 presents a tertiary sludge system using additional filtration.

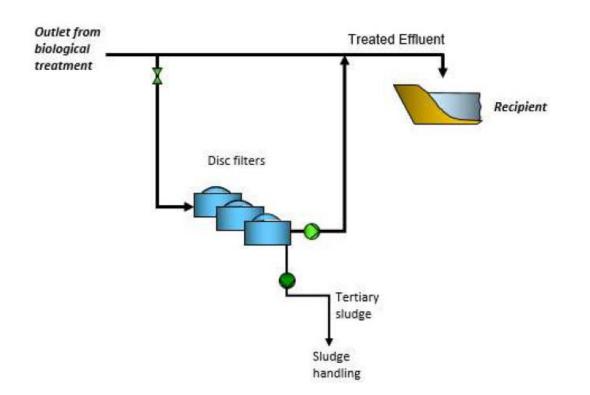


Figure 4.2. Tertiary sludge system using additional filtration. (Krogerus, 2016)

Because of hydraulic issues, several mills have installed disk type filters, e.g. Södra Mörrum, Södra Värö and UPM Changshu. Arauco Valdivia and Nueva Aldea use disks as tertiary stage.

Sludge from this type of treatment does not differ from biosludge.

#### 4.2.4 **Oxygenation** / ozonation

Tertiary sludge system using oxygenation is depicted in Figure 4.3. Adding chemical oxygen or ozone breaks more effectively the high molecular BOD (biological oxygen demand) still remaining in the wastewater.

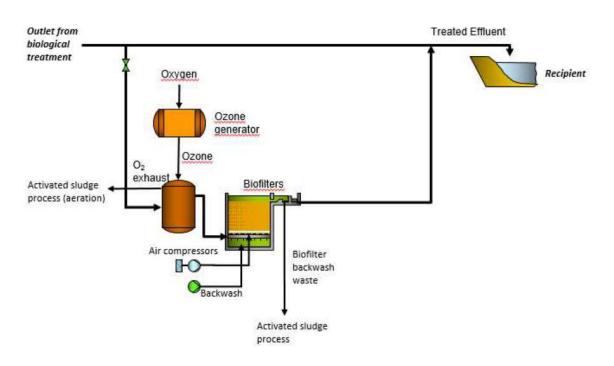


Figure 4.3. Tertiary sludge system using oxygenation. (Krogerus, 2016)

Only a few pulp and paper mills use oxygenation to improve wastewater quality. E.g. Laakirchen Papier, UPM Ettringen and Plattling, which all are paper mills in central Europe, use ozone.

#### 4.2.5 Metal removal by precipitation

Conventional wastewater treatment methods are geared towards removing organics that use oxygen (BOD), suspended solids (TSS), adsorbable organic halides (AOX), phosphorus and nitrogen. Metals are removed in conventional wastewater treatment with sludges to which they are adsorbed or taken up biologically. Some metals precipitate and are similarly removed with sludges. Due to high metals content a discharger may be required to implement additional metal control strategies to the pulp mill wastewater (Melcer et al., 1998).

Metals can be removed by precipitation using pH adjustment, carbonate precipitation or sulphide precipitation. The metal containing precipitate is not suited for mixing with other sludges.

cadmium. In Sweden Stora Enso Skoghall and BillerudKorsnäs Gruvön have been required to lower cadmium content in their wastewater (Sandström et al. 2016).

# 5 Sludge from raw water treatment

Both physical and chemical processes are used for raw water purification. They include sedimentation, coagulation, flocculation, filtration, and disinfection. Sludge is formed by the coagulation and flocculation. Usage of aluminium and iron salts are used for this purpose. Main problematic compounds are therefore aluminium sulphate and alum. The formed amount is highly dependent on the quality of source water, the quality and purity of alum and other chemicals used in the treatment (Feria-Díaz et al., 2016).

Water treatment can generate sludge from 0.5% to 3% of the total volume of treated raw water. Alum sludge disposal has been considered a problem because it is toxic to algae, fish, and other marine biota (George et al., 1995). Use of aluminium compounds is not desirable as soil improvers in agricultural land (Kupper et al., 2014).

	Feria-Díaz et al. (2016)	Franssila (2014)	Ahmad et al. (2016)
pН	5.1-8.0	3.8-4.1	6.9
Organics	64–140		
Al	27–153		80
Fe	5–37	60–170	32
Na	175		
Ν	4-5	0.2 - 1.0	
K	148		
Ash		800	890

Table 5.1. Example compositions of water treatment sludge dry matter (g/kg) based on references.

Wastewater sludge is filterable by centrifuge or vacuum filtration to dryness of 10-30%. Sludge in the backwash from freshwater treatment plant ~0.5 m<sup>3</sup>/ADt, containing <0.1 kg/ADt of material, has typically been either mixed with other wastewaters or dewatered and mixed with other sludges in wastewater treatment plant. If included into wastewaters, then most of the solids from freshwater treatment sludge are separated to primary sludge.

# 6 Typical problems with sludge utilization

The most often quoted problem with sludges is their high moisture content. When water content of sludge is high, the only impact of sludge incineration is to increase flue gas flow and losses without releasing additional heat in the furnace.

Sludges contain many substances, and some of them can be harmful. Odour from sludge causes typically only nuisance but can, if concentrated, be harmful to both health and air quality. Some components, such as heavy metals, can limit the disposal e.g. the usage as fertilizer. Additionally, biosludge contains substances, e.g. chlorine and alkali, which can cause problems in the incineration processes.

## 6.1 Corrosion problems

Fuel components can cause corrosion especially in boilers; the most problematic are chlorine and potassium. Chlorine itself can cause severe corrosion (Salmenoja & Mäkelä, 1999). Figure 6.1 shows an example of chlorine induced superheater corrosion. Alkali compounds, such as potassium, have a large impact on the corrosion rate. Biosludge in the forest industry includes chlorine because the incoming wood contains minerals and salts the wood has accumulated by drawing in water and evaporating it. Chlorine content in biosludge is higher than in traditional forest industry fuels; it can be assumed that biosludge incineration increases the corrosion risk. Part of the chlorine in biosludge is water-soluble and can therefore be removed using mechanical dewatering (Liimatainen, 2000).

Chlorine in fuel is partly devolatized during combustion forming either gaseous hydrogen chloride (HCl) or alkali chlorides, such as potassium chloride (KCl) and sodium chloride (NaCl). Alkali and metal chlorides reduce the melting temperature of boiler's deposits and consequently, the formed low melting compounds cause corrosion, especially in superheaters. Gaseous hydrogen chloride corrosion occurs typically in superheaters (Salmenoja, 2000).

Local conditions affect the severity of corrosion. According to Salmenoja (2000), the main factors are combustion temperature, local atmosphere in a boiler, and retention time in the furnace. In addition, materials used in the boiler affect the corrosion rate. For example, alloying chromium to steel typically increase resistance against corrosion. Compounds like sulphur, sodium, and potassium can accelerate the corrosion process (Anttikoski, 1992).



Figure 6.1. Example of chlorine induced superheater corrosion.

Corrosion risk is different for the bark boiler and the recovery boiler. The recovery boilers are designed for combusting black liquor that includes high amounts of challenging components like chlorine, potassium and sodium, and the combustion atmosphere and material selection are therefore suitable for challenging fuels. Thus, biosludge addition does usually not cause significant problems. Bark boilers are designed for firing bark or other wood residues, which do not contain as much harmful components as biosludge. Therefore, corrosion problems are more probable than in recovery boilers.

Superheater corrosion in bark boilers can be alleviated by using more expensive materials. E.g. in Imatra, after severe superheater corrosion was detected, the hottest superheaters were changed to X15 type material.

## 6.2 Fluidized bed agglomeration

Most of the modern biomass boilers in pulp mills are fluidized bed boilers. Effects of biosludge incineration on boiler emissions have been noted to cause bed agglomeration (Brus et al., 2005). Figure 6.2 demonstrates the problem.

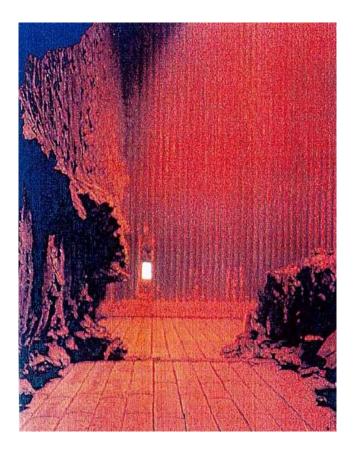


Figure 6.2. Example of fluidized bed agglomeration (Hulkkonen, 2006).

Typically, fluidized beds agglomeration shows as sudden defluidization of the bed, formation of hard substances and several days of unscheduled downtime. Typical wood

fuels exhibit coating-induced agglomeration. The cause can be low-melting calciumbased silicates (including minor amounts of, for example, potassium) or with high-alkalicontaining biomass fuels the formation of a layer of low-melting potassium silicate on the bed particles. When the number of contaminated particles gets high, they start sticking together and stop being fluidized. Increasing alkali and to a lesser degree phosphorus content in the incoming fuel increases agglomeration probability (Barišc et al., 2010).

#### 6.3 Effect on recovery boiler operation

Experiences of recovery boiler operation with and without biosludge mixing to black liquor have not observed large changes in operation (Mäntyniemi et al., 1995; Burelle et al., 2016; Haaga et al., 2017). Reduction efficiency, main emission levels and general boiler operation have typically remained unchanged as reported by mills using this method for the disposal of secondary sludge.

#### 6.4 Harmful emissions

Effects of biosludge incineration on biomass boiler emissions have been studied. Biosludge incineration does not increase the amount of sulphur dioxide, heavy metals, PAH-compounds, or incombustible materials (Liimatainen, 2000).

Biosludge incineration, however, increases both recovery boiler and biomass boiler NOx (Suhr et al., 2015; Lecomte et al., 2017) because biosludge contains significant amount of nitrogen, then fuel nitrogen loading of the boiler increases. As, in general, about one third of the nitrogen in fuel is converted to NOx in the lower furnace, the increase in incoming fuel nitrogen explains the trend.

Biosludge behavior in evaporators has been studied (Vähä-Savo et al., 2012). Only a small portion of nitrogen in biosludge was found to be released during evaporation. Most of the biosludge-N ended up with black liquor to the recovery boiler. Clear increase in recovery boiler NOx has not been measured, but single droplet experiments record 120–140% increase in NOx.

Välttilä et al., (1994) studied biosludge incineration in a BFB, CFB and grate boiler and measured the effect on emissions. Biosludge contains chlorine and especially organic chlorine compounds. It is notable (Table 6.1) that significant increases of chlorinated dioxins and furans, polychlorinated biphenyls (PCBs) as well as chlorinated phenols and benzenes were observed. The origins of dioxins are most probably at least partly caused by higher dioxin and phenol content in the burnt biosludge.

Table 6.1. Flue gas emissions when incinerating bark, biosludge and auxiliary fuel. (Välttilä et al., 1994)

Boiler		B	FB, Trial	1		CFB			Grate	
Fuel mix		Bark	Bark sludge	Bark sludge gas	Bark	Bark sludge	Bark sludge coal	Bark	Bark sludge	Bark sludge peat
Boiler		-		0						pour
Output	MW	72	65	83	62	59	61	49	47	55
Bed temp.	°C	865	845	855	905	840	900			
Furnace temp.	°C	875	830	880	920	900	950	925	900	725
ESP temp.	°C	145	150	150	320	330	320	155	155	150
Fuel **										
Mixed sludge	%		8.7	8.2		13.9	19.5		6.5	5.6
Moisture	%	52	57	56	55	60	58	57	58	55
Chlorine	mg/kg	250	1340	1310	120	570	520	110	1000	580
PCDD/PCDF, Eadon	pg/g	5.9	99	78	<5	1.0	1.8	2	12	13
Aux. fuel, of heat input	%			25			20			15
Flue gas, reduced to 10% oxy	zen content									
Volume	m <sup>3</sup> n/s	43	40	48	41	41	41	33	32	37
со	mg/m <sup>3</sup> n	25	37	5	450	800	160	260	210	180
SO <sub>2</sub>	mg/m <sup>3</sup> n	26	26	25	29	21	26	17	36	49
$NO_x$ (as $NO_2$ )	mg/m <sup>3</sup> n	260	300	310	250	270	280	160	200	230
Particulate	mg/m <sup>3</sup> n	38	20	20	50	110	45	180	210	260
HCI	mg/m <sup>3</sup> n	2	5	34	2	2	5	4	5	15
Cl-phenols	µg/m³n	-			1.61	1.9	2.6	0.51	0.24	0.48
Cl-benzenes	$\mu g/m^3 n$	0.08	0.56	0.11	0.34	0.43	1.37	0.4	0.67	0.55
PCBs	$\mu g/m^3n$	0.04	-	-	0.32	0.63	0.63	0.27	0.30	0.05
PAHs	μg/m³n	4.1	4.6	6.4	1370	850	450	8.5	6.6	1.1
PCDD/PCDF, Eadon	r.a. ii					200		0.0	5.0	
Stack	ng/m³n	0.04	0.14	0.07	0.39	0.71	1.21	0.09	1.09	0.91
Before ESP	ng/m³n				0.04	0.15	0.43 *			

Exceptionally high value of 1.8 ng/m<sup>3</sup>n in parallel sample disregarded.

\*\* Contents are those for dry bark-sludge mixture.

Black liquor contains both elemental chlorine and organic chlorine compounds. Most probably due to catalytic reactions, the resulting chlorinated dioxins and furans, PCBs as well as chlorinated phenols and benzenes have been observed only at very low, barely measurable quantities. Harila and Kivilinna (1998), measured <10 pg/m<sup>3</sup>n of PCDD as

TCDD equivalent (i.e. below detectable limit) for both black liquor mixed with biosludge and black liquor without biosludge.

#### 6.5 **Problems with non-process elements**

Biosludge addition to the recovery cycle has been reported as increasing contents of magnesium, aluminium, silicon, and phosphorus (Bialik et al., 2014).

The components of biosludge can cause problems also in other forest industry process stages than the combustion processes. Non-process elements (NPE) can exit from forest products mill through wastewater, dregs, grits, biomass boiler ash, lime mud, or recovery boiler precipitator (ESP) ash.

## 6.5.1 Effect on evaporator fouling

NPE in biosludge can cause build-up in the evaporator heat transfer surfaces. When biosludge is fed in the evaporators, aluminium or zinc can form harmful deposits, e.g. aluminium forms sodium aluminium silicates. Increasing flow of other NPEs can either concentrate on evaporators and cause fouling or cause problems in causticizing.

#### 6.5.2 Effect on lime mud dry solids

Lime mud from white liquor separation is filtered and then dried using flue gases from the lime kiln. The dryer is a mixing chamber where lime mud contacts with the flue gas. Using the lime mud dryer, lime kiln capacity can be increased. Mud from the dryer is separated from the flue gas in a cyclone and fed into the lime kiln, see Figure 6.3. The calcination process, where  $CaCO_3$  reacts to form CaO and  $CO_2$ , occurs in the lime kiln at the temperature of  $1100^{\circ}C$ . The required heat for the calcination process is generated by burning fuel; usually natural gas when it is available or heavy fuel oil is used. In some mills, fossil fuels are replaced with biogas produced from waste wood and bark in the gasifier. After burning, the lime is cooled in a sector cooler where heat is recovered to heat up the combustion air. As phosphorus is enriched in ESP dust, then purging ESP dust will remove phosphorus effectively.

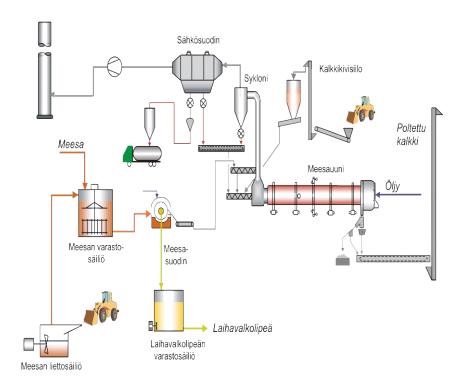


Figure 6.3. Lime kiln in a modern kraft pulp mill (Pöyry, 2007).

Phosphorus in sludge can enrich into lime cycle and cause lower lime mud dry solids. This creates the need for additional make-up lime and requires dumping of more lime mud (Valmet, 2016). Makeup lime at the rate of 2 to 5 kg/ADT of pulp was needed in one case to control phosphorus accumulation (Burelle et al., 2016).

# 7 Sludge disposal

Sludge disposal has always been challenging. Tightening legislation and the demand to stop sludge disposal as solid waste have made it even more difficult. Traditionally, sludges have been incinerated in forest industry boilers or disposed of to landfills. In 2014, over 60% of generated sludge in the Finnish forest industry was used for energy production (Alakangas et al., 2016). The main challenge is the low or even negative heating value. Most commonly sludges are incinerated mixed and in bark boilers on site after mechanical drying (Ojanen, 2001; Strömberg, & Herstad Svärd, 2012). Biosludge is sometimes fired separately in recovery boilers. Less than half of sludge ends up in landfills or is utilized. The fibre-rich primary sludge has more usage possibilities than other sludges and is also more valuable as fuel. The heterogeneous properties of sludges complicate their use and disposal. The properties vary even inside the mills depending on the products, processes, and chosen technologies. Of the biosludge generated in Sweden, more than half was incinerated, and the rest was used for land improvement, as cover material and composted (Fuglesang et al., 2015).

Two directives control waste management in the European Union (EU): directive 2008/98/EC on waste and directive 31/1999/EC on the landfill of waste. In addition, countries have their own regulations that control waste management (Kostamo, 2016). Landfilling of biodegradable wastes is prohibited in the EU. The directive on waste orders primarily to prevent the generation of waste. All waste generation cannot be prevented and therefore, there is also regulations for waste disposal. The desired order for waste disposal is the following: preparing for re-use, recycling the materials, energy recovery or utilization in other ways, and finally, landfill disposal (Figure 7.1). According to the directive, it is recommendable to use sludge firstly as a raw material and only secondly as an energy source.

The main problem in biosludge disposal is its high moisture content. The moisture content of mechanically dewatered biosludge is at or below 10–15% and further drying is



challenging due to the structure of sludge. Biosludge disposal is costly unless water can be removed with moderate effort and energy consumption.

Figure 7.1. Waste manage hierarchy according to the directive 2008/98/EC on waste (European Commission, 2016).

Several attempts have been made to find feasible disposal methods for biosludge. Researchers have studied both more efficient dewatering methods and disposal alternatives. The most widely used disposal processes are incineration in a bark boiler or a recovery boiler, and the most promising new alternatives are hydrothermal carbonization (HTC) and biogas production.

In Finland, the forest industry is an important industrial sector. There are 19 pulp mills, 17 paper mills and 14 paperboard mills (Finnish Forest Industries, 2018). Many mills utilize active sludge process in wastewater treatment plants and therefore, produce biosludge. The mills use various methods for sludge disposal. This section introduces examples on biosludge treatment processes in Finnish mills. The example mill cases Äänekoski, Kemi, and Kuopio highlight the newest sludge management as stated in environmental permit applications (see section 7.2). An older Imatra case highlights a typical treatment option, where, biosludge is incinerated in a bark boiler (Välttilä et al., 1994). Lastly in Heinola fluting mill, a new HTC process, which treats biosludge is planned to start in 2020.

## 7.1 EU BAT BREF

The EU Best Available Techniques (BAT) reference documents (BREFs) for various industrial sectors give information on the techniques and processes considered the best in terms of emissions and consumption levels. According to Suhr et al. (2015), sludge handling consists of two main stages; firstly, thickening and dewatering and secondly, disposal. Typically, biosludge is mixed with primary sludge to improve the dewatering properties, but sometimes pure biosludge is handled. Different mechanical dewatering methods are used to decrease volume and weight and to increase dry solids content. Decanter centrifuge is usually used for pure biosludge. Dewatering decreases transportation costs and makes disposal easier. Biosludge is usually pre-dewatered before the actual dewatering process using gravity tables, gravity and hydrostatic disc thickeners, and thickener centrifuges. Dry solid content increases then from 1–2% to 3–4%. Dry solid content of mixed sludge can be at 25–35% after belt or filter presses and 40–50% after a screw press using steam.

Excess thermal energy can be used for drying the sludge, when excess heat is available, and the benefits exceed the expense of the investment. Drying decreases the need for supportive fuels in the incineration of sludge. Fluidized bed boilers are the most suitable boilers for sludge incineration. Boilers require 35–40% dry solid content for spontaneous burning. Biosludge can reach dry solid content of 18–22%, and thus, biosludge always needs support fuels. Chemical sludge cannot be incinerated without auxiliary fuels due to high content of inorganic material and water.

Table 7.1 presents emissions of a fluidized bed boiler incinerating biosludge, deinking sludge, biogas, natural gas, and refuse-derived fuel. All measured emissions are lower than the limit values. Besides incineration, biosludge can be used as a fertilizer if it does not contain harmful components. Primary sludge is used for example in brick industry. Separate handling makes the further use of sludges easier.

Parameter	Measured values in 2008 <sup>1</sup> [mg/Nm <sup>3</sup> ]	Limit value daily average <sup>1</sup> [mg/Nm <sup>3</sup> ]	Installed emission abatement techniques
Dust	5	10.0	Bag filters (Teflon)
SO <sub>2</sub>	1	50.0	S content in the sludge is very low
NO <sub>X</sub>	190	200.0	SNCR (injection of ammonia)
NH <sub>3</sub>	Not detectable	20.0	No relevant ammonia slip
СО	2	50.0	Fluidised bed ensures good combustion
HCI	3	10.0	Injection of an adsorbent (mixture of calcium and activated carbon) before the bag filters
Total-C	1	10.0	

Table 7.1. Emissions of a fluidized bed boiler burning biosludge, deinking sludge, biogas, natural gas and refuse-derived fuel. (Suhr et al., 2015)

NB: Heavy metals and dioxins are well below the limit values. They are measured periodically, e.g. Hg <0.01 mg/m<sup>3</sup>; PCDD/F: maximum value: <0.004 ng/m<sup>3</sup>.

<sup>1</sup>The measured values and the emission limits refer to an oxygen content of 11% per volume and are calculated as yearly average values.

Source: Landesdirektion Leipzig, Abt. Umwelt, Stora Enso Sachsen: Continuous emission monitoring. Yearly emission report 2008

## 7.2 Sludge disposal in the newest Finnish cases

The newest Finnish pulp mill Metsä Fibre Äänekoski started in 2017. This 1.3-millionton mill is the largest single-line pulp mill in the northern hemisphere (IRENA, 2018). Finnpulp Oy applied for an environmental permit to a greenfield Kuopio Bioproduct mill. Even though the permit was initially given, on December 19, 2019, the Supreme Administrative Court (KHO) in Finland rejected the environmental permit application. Metsä Fibre Oy has applied for an environmental permit to Kemi Bioproduct mill. At the time of writing, the environmental permit application of Kemi is still awaiting first decision from the Regional State Administrative Agency. The Äänekoski mill produces biogas from biosludge in an anaerobic digestion process. The Kemi pulp mill currently incinerates biosludge in the recovery boiler.

### 7.2.1 Disposal at Metsä Fibre Äänekoski bioproduct mill

Metsä Fibre states in its application (Metsä Fibre 2014) that a biogas generating process step will be included. This process step will use the 11–12 kg/ADt of primary-, bio- and

tertiary sludge generated. This is one of the first commercial applications of biogas from sludge at a pulp mill.

"Laitoskokonaisuus sisältää sellutehtaan lisäksi tuotekaasulaitoksen, jossa prosessin sivutuotteina syntyvää biomassaa jalostetaan tuotekaasuksi ja mädättämön, jossa käsitellään jätevedenpuhdistamon liete."

Because of the newness of biogas generation, the permit application later includes the additional possibilities to burn sludges in the recovery boiler or to gasify them. Primary sludge can also be incinerated together with bark or composted, and tertiary sludge can be incinerated in the lime kiln.

The biological wastewater treatment plant of the mill will also handle the wastewaters of other business entities operating at the same site; Speciality Minerals Nordic Oy PCC-plant, CP Kelco Oy CMC-plant and Metsä Board Oy board mill as well as from Äänekoski community. The wastewater plant comprises of primary clarifier, cooling of wastewater in a cooling tower, a new MBBR-plant (moving bed biofilm reactor) with treatment continuing in an old wastewater plant anaerobic treatment followed by tertiary phase with flotation.

"Uudistettu biologinen jätevedenpuhdistamo tulee käsittämään kiintoainepitoisten jätevesien esiselkeytyksen, jäteveden jäähdytyksen, kantoaineprosessin (MBBR), neutraloinnin ja ravinteiden syötön, ilmastuksen, tertiäärivaiheen, jälkiselkeytyksen sekä palautus- ja ylijäämälietteen käsittelyn. Kantoaineprosesissa (MBBR), jonka toiminta perustuu biofilmiin, mikrobikasvusto tapahtuu kantoainekappaleiden pinnalla aerobisissa olosuhteissa. MBBR-prosessin jälkeen tarvitaan vaikeammin hajoavan kemiallisesti happea kuluttavien aineiden (COD) hajottamiseksi aktiivilietelaitos ja tertiääriflotaatio."

All sludges from MBBR and the old secondary treatment are handled together.

In the permit an alternative to primary  $\sim 3 \text{ kg/ADt}$  and tertiary  $\sim 4 \text{ kg/ADt}$  sludge utilization is either to mix it with bark before the dryer and to gasify bark/sludge mixture or to mix primary sludge with biosludge. To enable an efficient gasification the sludge needs to be with the bark.

"Primääriliete syntyy jätevedenpuhdistamon esiselkeytyksessä, jossa laskeutetaan jäteveden kiintoaine. Tämä jae sisältää kuitu-, kuori-, täyte- ja lisäaineita sekä pigmenttejä. Syntyvä primääriliete kuivataan kartioruuvipuristimilla ja poltetaan kuoren mukana meesauunilla."

In the permit the main alternative of bio- and tertiary sludge 8 kg/ADt utilization is to make biogas from it, use the gas and dispose of digestate outside the mill.

"Biologisella jätevedenpuhdistamolla syntyvä kuitu- ja bioliete käsitellään mädättämöllä."

The used biogas process comprises of pretreatment, bioreactors, biogas storage, carbon dioxide separation, mechanical dewatering and thermal drying, Figure 7.2.

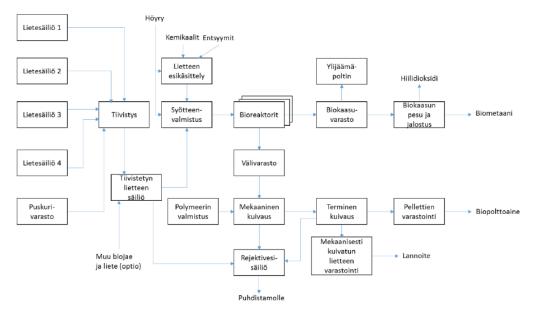


Figure 7.2. Processes in the biogas plant (EcoEnergy SF, 2017).

According to its environmental permit, Metsä Fibre Äänekoski bioproduct mill (1 300 000 ADt/a pulp) plans to

- Dispose 15 000 t/a of sludges from wastewater treatment at biogas production plant. About 40% of the sludges is from the tertiary treatment. As an alternative, they propose burning it at recovery boiler.
- They propose as option drying and disposal of primary and tertiary sludges at lime kiln gasifier.
- 3. Has not considered where the digestate from biogas plant will be disposed at.

## 7.2.2 Disposal at Finnpulp Kuopio

The unsuccessful environmental application by Finnpulp Oy did originally apply for traditional wastewater treatment. In additional submission it did take into account possible tertiary treatment in its wastewater plant (Krogerus 2016). Figure 7.3 presents the byproduct flows in the Finnpulp mill. The environmental application for Finnpulp (2016), states

"Tertiääripuhdistuksella voidaan vähentää erityisesti vesistöön kohdistuvaa happea kuluttavan aineen (COD<sub>Cr</sub>) ja fosforin kuormitusta. Tertiäärikäsittely vaatii kemikaalien käyttöä, mistä johtuen erityisesti sulfaattikuormitus vesistöön kasvaa merkittävästi. Myös natriumin kuormitus lisääntyy, mutta vähäisemmässä määrin. Kemikaalit sisältävät myös alumiinia, joten alumiinin kuormitus lisääntyy."

which can be translated as "Using tertiary treatment reduces especially the oxygen demand ( $COD_{Cr}$ ) and phosphorus load to recipient waters. Tertiary treatment requires chemicals, which increases especially the sulphate load. In addition, the sodium load increases, but to a lesser extent. The chemicals contain aluminium, so aluminium loading increases." The sulphate loading increase was estimated as +42%.

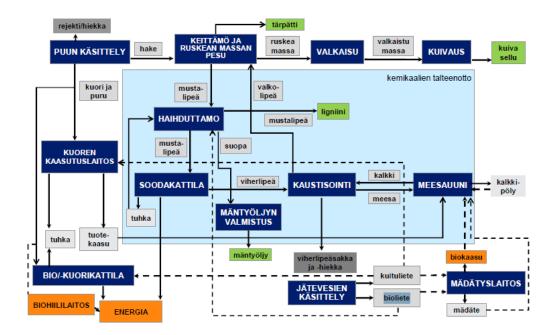


Figure 7.3. Flows of byproducts in Finnpulp (Finnpulp, 2016).

The mill estimated that using tertiary treatment one could reduce the chemical oxygen demand from 33 t/d to 19.5 t/d (~40%). Biological oxygen demand reduction was lower, from 1 425 kg/d to 1 170 kg/d (~20%). Biological oxygen consuming matter breaks effectively already in the normal secondary stage.

According to its environmental permit application, Finnpulp Oy (1 200 000 ADt/a softwood pulp) plans to

- Dispose 10–12 000 t/a primary sludge (kuituliete) at biogas production plant (mädätyslaitos) and or at biomass boiler (bio/-kuorikattila). As an alternative, they propose drying and disposal at lime kiln gasifier (meesauuni).
- Dispose 8–9 000 t/a secondary sludge (bioliete) after it has been centrifuged at evaporator (haihduttamo) to be combusted in the recovery boiler mixed into the black liquor.
- 3. Has not considered where specifically the tertiary sludge will be disposed at.

## 7.2.3 Disposal at Metsä Fibre Kemi bioproduct mill

According to the environmental permit application of Metsä Fibre Oy (2019b), the expansion of old mill to new 1 500 000 ADt/a soft-/hardwood pulp, plans to use current (existing) wastewater treatment with possible option of tertiary treatment. They plan to be able to keep the current COD loading at <40 t/d.

- Dispose 5 000 t/a primary sludge by burning in the current biomass boiler and or composting.
- Dispose 10 000 t/a secondary sludge to be incinerated in the recovery boiler mixed into the black liquor. As an alternative they propose anaerobic treatment in biogas plant.
- 3. Has not considered where specifically the tertiary sludge will be disposed at.

Solid fraction from biogas plant is not considered hygienically suitable for farm usage.

## 7.3 Sludge disposal by burning it in recovery boiler

Incineration in a recovery boiler is a potential way to dispose of biosludge. Primary and tertiary sludges have not been disposed of at recovery boilers. Biosludge burning has not caused large problems because recovery boilers are designed for challenging fuels. Additionally, they have advanced monitoring and control systems, which is very helpful when biosludge incineration is considered. The share of biosludge in black liquor is low and consequently, impact on black liquor properties is moderate. For example, Kemi pulp mill has incinerated biosludge in the recovery boiler since 1993 without reporting associated problems (Harila & Kivilinna, 1999).

In the recovery boiler the organics of sludge combust. Some of the NPE, notably Cl, is vaporized and found from ESP dust. Most inorganics from sludges continue with smelt to causticizing.

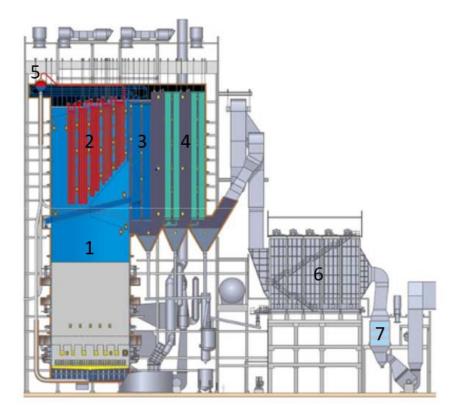


Figure 7.4. Modern recovery boiler; 1. Furnace 2. Superheaters 3. Boiler banks 4. Economizers 5. Steam drum 6. Electrostatic precipitator 7. Heat recovery system, courtesy of Valmet.

#### 7.3.1 Causticizing and lime kiln

The purpose of causticizing is to convert green liquor to white liquor. Sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) in green liquor is converted to sodium hydroxide (NaOH) using calcium oxide or lime (CaO). The process in depicted in Figure 7.5.

Green liquor is pumped to a slaker where lime reacts with water in the green liquor forming slaked lime (Ca(OH)<sub>2</sub>). The reaction produces heat. The causticizing reaction continues in the causticizing tanks. Ca(OH)<sub>2</sub> and Na<sub>2</sub>CO<sub>3</sub> react to produce sodium hydroxide (NaOH) and calcium carbonate (CaCO<sub>3</sub>), also called lime mud. After the causticizing process, white liquor is pumped to white liquor separation, where white liquor and lime mud are filtered or clarified.

NPE from sludge is mostly removed with other undesirables in green liquor filtration. Notable exception is phosphorus that enriches in lime cycle. Phosphorus causes low filtration lowering the lime mud dry solids and kiln capacity.

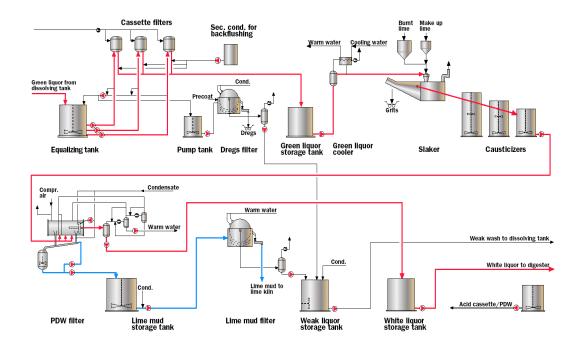


Figure 7.5. Modern white liquor preparation (Löwnertz, 2005).

#### 7.3.2 Sludge handling for incineration in recovery boiler

When biosludge is incinerated in the recovery boiler, it is usually mixed with black liquor in the evaporation plant. The sludge needs to be fed into the higher dry solids effects to prevent NPE in it to spread into all effects. Sludge containing liquor is pumped directly from evaporator to the pressurized liquor tank and then from tank to be burned in the furnace of the recovery boiler. Sludge behaviour at evaporator viz deposit formation and fouling during evaporation needs to be considered carefully.

A possible disposal method is shown in Figure 7.6. Biosludge is mixed with intermediate/strong black liquor at the evaporators and the mixture continues to the recovery boiler for incineration. Usually, the share of biosludge dry solids in black liquor is about 1%. Often the dry solid content of biosludge is raised before the evaporators using additional polymers. The polymers enhance dewatering properties of the sludge and promote flocculation. Sludge treated with polymers is delivered to actual dewatering unit where dry solid content increases from 0.5–3.0% to 8–12%. Typically, decanter centrifuge is utilized for dewatering. Without sludge drying, the load at the evaporators increases undesirably (Valmet, 2016). Increased load increases steam consumption and reduces the dry solids capacity of the evaporators.

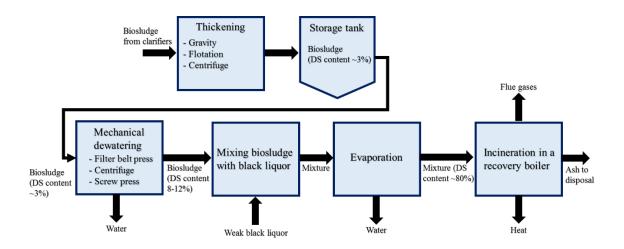


Figure 7.6. Biosludge disposal in a recovery boiler. (Valmet, 2016)

According to Harila & Kivilinna (1999), seven important factors need to be taken into account, when recovery boiler is used for biosludge disposal. The factors are biosludge pre-treatment including mainly dewatering, behaviour of biosludge during evaporation, effects of emissions on the environment, changes in combustion properties, economy, corrosion, and other risks. In addition, NPE in biosludge and their behaviour and accumulation in the equipment and in the recovery cycle have to be taken into account.

#### 7.3.3 Case Kemi

Metsä Fibre Kemi is a pulp mill located in Northern Finland. The current mill capacity is 620 000 tons of pulp per year (Metsä Fibre, 2019b). In 2006, Kemi pulp mill produced 38 390 tons biosludge with dry solid content of 10% (Pohjois-Suomen ympäristölupavirasto, 2007). The mill has disposed of biosludge by incinerating it in a recovery boiler since 1993. Biosludge is dewatered to up to 8–12% dry solid content using a decanter centrifuge with presence of polymers. Dewatered sludge is mixed with weak black liquor and the mixture is fed into the evaporation plant. The dry solid content is raised, and then the mixture is incinerated in the recovery boiler (Valmet, 2016). The process is presented in Figure 7.7.

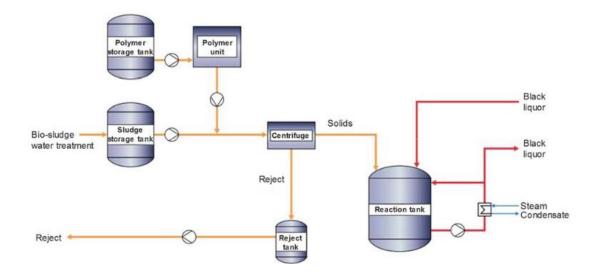


Figure 7.7. Biosludge incineration in the recovery boiler in Kemi mill (Harila & Kivilinna, 1999).

The share of biosludge is about 0.5% of the amount of black liquor in the Kemi mill. Low sludge concentration leads to only a moderate effect on the recovery boiler's operation. The mill operators have not noticed significant changes in the process parameters after installing the biosludge disposal system. Combustion properties have changed only little, chloride content has not increased, non-process element accumulation points or fouling in the evaporator have not been noticed, and no noticeable effects on the emissions to air have been seen. Viscosity of the fluid has varied slightly, and the heating value has even improved after biosludge addition (Harila & Kivilinna, 1999).

## 7.4 Sludge disposal by incineration in bark boilers

Many forest industry mills have a bark boiler or a power boiler to fire residual biomass and to generate heat. Bark boilers are usually bubbling fluidized bed (BFB) or circulating fluidized bed (CFB) boilers. Both boilers operate using solid sand particle bed that is in fluid-like state because of the primary air flow through it. These boilers are suitable for biofuels with a high moisture content because the bed has a high heat capacity. Until recently, also grate boilers were utilized for incinerating bark and other wood residues in the forest industry. Incineration in a bark boiler is the main disposal method for primary sludge and often also for biosludge.

#### 7.4.1 Sludge handling for burning in bark boiler

Typically, sludge incineration in a bark boiler consists of several stages (Figure 7.8). Firstly, sludge is dried to reach as high dry solid content as possible with reasonable amount of effort. Dried sludge is mixed with bark and possibly also with other wood residues. Finally, the mixture is incinerated in a bark boiler. The share of sludge can be about 5–10% of the mass flow of bark. Incineration in a bark boiler requires dry solid content of 35–45% if supportive fuels are not used. The heating value as fired should be at least 5 MJ/kg. In the case of biosludge, supportive fuel is practically always needed (Ojanen, 2001). Usage of supportive fuel, such as natural gas, increases costs.

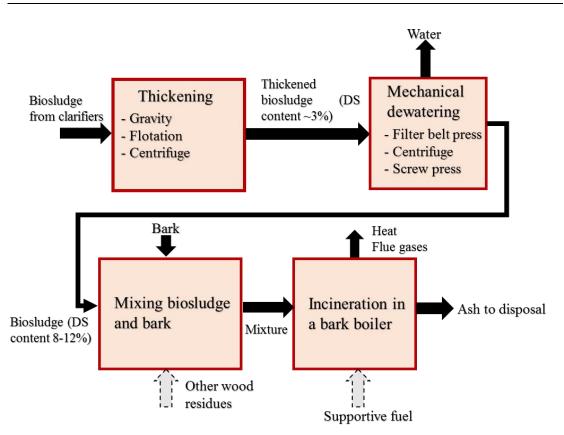


Figure 7.8. Biosludge disposal by incinerating in a bark boiler.

Sludge incineration in a bark boiler is a widely used disposal method. Incineration is an attractive process because it significantly decreases the volume of sludge to be landfilled and reduces costs caused by other disposal methods. The challenge is the high moisture content, especially in the case of biosludge. Amount of heat released by incinerating sludge is low or even negative.

In pulp mill bark boilers, the main fuel is sometimes also wet and incineration can be difficult even without biosludge. In addition, if sludge cannot be fed into boiler in wintertime, it will freeze in piles. Frozen sludge can, however, be used during the following summer.

Sludge disposal is desired to be economical. Sludge incineration causes some additional costs compared with boiler's normal operation. The cost is related to the moisture content. The additional cost consists of energy that is required to vaporise the water, boiler losses,

increased amount of flue gas, heat that exits with hot ash, ash transportation, treatment costs, and increased need for maintenance. In addition, the level of energy prices has an effect on the costs (Lehtinen, 2001).

### 7.4.2 Case Imatra

Stora Enso Imatra mill is a large pulp mill producing sulphate pulp, CTMP, and paperboard. The capacities of both pulp and paperboard are over 1 000 000 tons per year (Stora Enso, 2019b). Imatra mill disposes of all sludges by incinerating them in a bark boiler. The sludges are mixed and then dried using screw press with polymers and steam before incineration (Itä-Suomen ympäristölupavirasto, 2007). About 40 000 tds of sludge is incinerated annually. In 2003, the share of biosludge was about 6% of the mixture of bark and sludge.

## 7.5 Torrefaction and hydrothermal carbonization of sludge

Torrefaction is a biomass treatment process that occurs at a temperature range of 200– 300 °C and in atmospheric pressure, typically in the absence of oxygen (Basu, 2013). The residence time is relatively long, typically hours. The operating temperature is limited below 300 °C because higher temperatures are harmful due to devolatilization and carbonization of polymers. Also, loss of lignin and thermal cracking of cellulose accelerate when temperatures rise above 300 °C. The process produces torrefied biomass that is coal-like material. The operating principle of the torrefaction process is presented in Figure 7.9. About 70% of biomass feed is converted to torrefied biomass and 30% to volatiles and gases. Torrefaction decreases the energy content of the feed by approximately 10% but significantly increases its energy density.

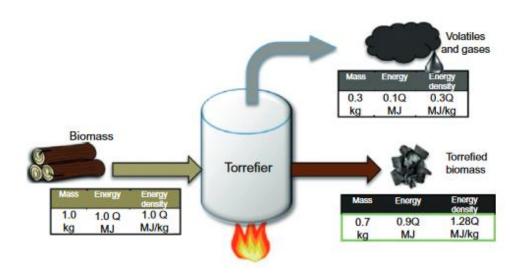


Figure 7.9. The torrefaction process. (Basu, 2013)

Dry torrefaction is, however, unlikely to be feasible when the moisture content of the feed material is high, above 50–70% (Libra et al., 2011). Hydrothermal carbonization (HTC) is a variation of traditional torrefaction and more suitable for wet biomass. HTC is a thermochemical process where biomass is treated in a suspension of water to produce solid, coal-like product that can be called hydrochar. Additionally, liquid and gaseous byproducts are created. Yields of solid, liquid, and gaseous products are in the ranges of 50–80%, 5–20%, and 2–5%, respectively (Libra et al., 2011).

### 7.5.1 Sludge handling for making biochar in HTC

HTC occurs in a closed system at a temperature range of 180–250 °C and under autogenous pressure. Residence time is typically several hours. Various types of biomass have been successfully treated with HTC to produce fuel that is stable, non-toxic, and easier to handle than the original biomass (Funke & Ziegler, 2010; Hirvelä, 2018). Produced hydrochar can be used for several purposes, such as replacing coal in energy production applications. A possible flow chart of the HTC process is presented in Figure 7.10.

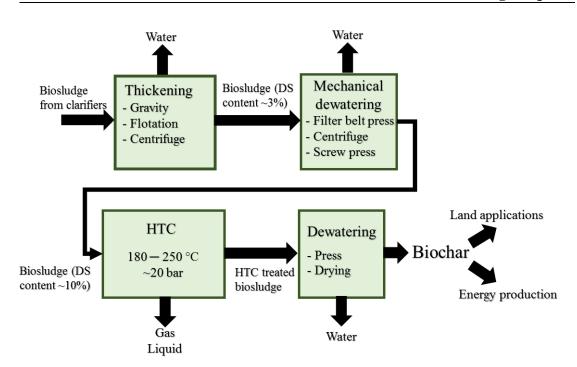


Figure 7.10. HTC process.

HTC of biosludge is an attractive concept for several reasons. Firstly, technoeconomically feasible and environmental-friendly treatment processes for biosludge are greatly needed. Secondly, HTC treatment can be applied even when sludge has a high moisture content and no preceding, energy-intensive drying is needed (Alatalo et al., 2013). Thirdly, the high heating value of biosludge solids is a benefit when HTC treatment is considered (Child, 2014). HTC also destroys organic pollutants and produces a sterile end-product that can be used not only as an energy source, but also as e.g. soil conditioner (Funke & Ziegler, 2010; Libra et al., 2011).

HTC treatment raises content of carbon in biosludge but decreases the contents of oxygen, hydrogen and nitrogen (Zhang et al., 2014). The heating value after the process is slightly higher than the heating value of the raw material. The heating value of dry biosludge solids after HTC is above 20 MJ/kg while the heating value of fossil coal is around 30 MJ/kg. First industrial-scale biomass HTC plant was introduced in 2010 by AVA-CO<sub>2</sub> (Silakova, 2018). The technology is developing and not widely used.

#### 7.5.2 Case Heinola

Stora Enso Heinola mill is located in Southern Finland. It produces fluting from semichemical pulp 300 000 tons in year (Stora Enso, 2019a). Biosludge has been combusted in a fluidized bed boiler (Ojanperä, 2019). At the time of writing, Heinola mill is starting HTC treatment of biosludge to produce usable biofuel in co-operation with C-Green Technology AB. Figure 7.11 presents the HTC process. The pressurized HTC process uses heat and produces hydrochar (Axegård, 2019). Hydrochar will be used in the mill to reduce the use of fossil fuels and production of fossil-based CO<sub>2</sub> emissions. 16 000 ton of biosludge can be treated into solid biofuel annually using the new process (Ojanperä, 2019).

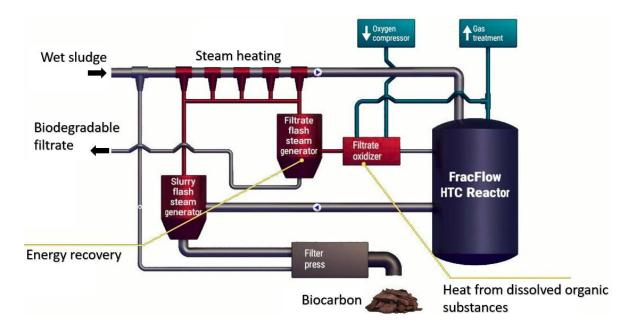


Figure 7.11. C-Green HTC process for biosludge (Modified from Axegård, 2019).

### 7.6 **Biogas generation**

Sludges can be treated in an anaerobic digestion process. This has several advantages. It reduces the volume and dry mass of sludge and produces utilizable biogas as a byproduct. Digestion process has a low energy consumption. Biogas can be used for energy production and the digested sludge, digestate, can be used for land applications or incinerated. Dewatering the digestate can be more difficult than the original sludge, which

can complicate its incineration. Typically, problem is solved by mixing primary sludge or other biomaterial like grass with the digestate. Digestate also is suitable for soil improvement in other than food production lands (Hagelqvist, 2013).

Anaerobic treatment of industrial wastewaters including biogas production has been widely used in the pulp and paper industry since the early 1980's (Habets & Driessen, 2007), but typically it is only applied to few selected wastewater streams (Meyer & Edwards, 2014). Anaerobic treatment often precedes aerobic activated treatment plant and then decreases biosludge yield (Habets et al., 2002). Methods for biogas production from wastewater have interested investors also lately. For example in Estonia, a new biogas plant commissioned in 2014 produces one third of the energy requirement of the pulp mill (Dutch Water Sector, 2017), and a paper mill in Sweden is constructing a plant for production of liquified biogas for transportation use (Gasum, 2020). However, biogas generation from forest industry sludges using anaerobic digestion is a novel process (Meyer & Edwards, 2014) and so far, has been applied only to biosludge in the forest industry.

#### 7.6.1 Sludge handling for making biogas

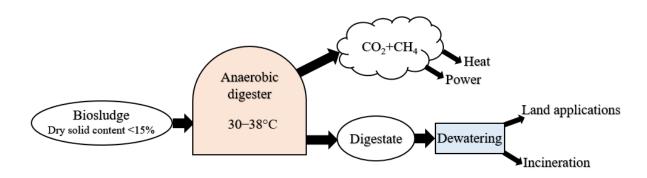
Anaerobic digestion process occurs in the absence of oxygen. The temperature is usually in the mesophilic area (30–38 °C). Organic material in biosludge decomposes forming biogas. Desired gaseous product is methane (CH<sub>4</sub>) that can be effectively used for energy production. Depending on the used technology, about 55–75% of produced biogas is methane and the rest is mainly carbon dioxide (CO<sub>2</sub>) (Liimatainen, 2000). The following equation presents the biogas forming process (Tchobanoglous et al., 1993):

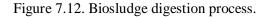
$$C_aH_bO_cN_d \rightarrow nC_wH_xO_vN_z + mCH_4 + sCO_2 + rH_2O + (d - nx)NH_3$$
(2)

In the equation, s = a-nw-m and r = c-ny-2s.

Produced biogas can be fired in a boiler or with a gas burner for heat and electricity production. The heating value of the biogas is about 19–23 MJ/m<sup>3</sup>. For comparison, the

heating value of natural gas is about 36 MJ/m<sup>3</sup> (Alakangas et al, 2016). A simplified biosludge digestion process is presented in Figure 7.12.





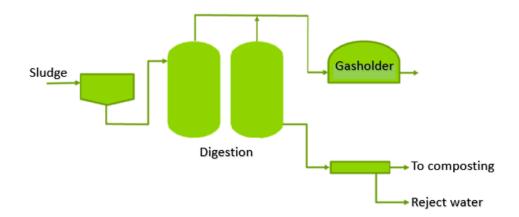
Most studies on biosludge digestion discuss municipal sludge or sludge from varying industrial sources, but not often from pulp and paper industry (Meyer & Edwards, 2014). The properties of forest industry sludges differ from the others especially due to high content of lignocellulosic material, which harms the digestibility of sludge. Pure biosludge digestion leads to lower yield than for digestion of for example municipal sewage sludge. The yield can be improved for example by mixing forest industry sludge with municipal sewage sludge (Hagelqvist, 2013), or other feedstocks with appropriate properties, considering e.g. feedstock biodegradability, total solid content, and C/N ratio (Veluchamy & Kalamdhad, 2017). The methane yield of untreated sludge varies widely based on sludge properties and digestibility; pretreatment, such as thermal or microwave treatment, can improve the yield (Meyer & Edwards, 2014).

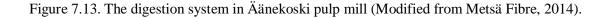
Biogas production from biosludge requires external heating. The process is exothermal, but the reaction heat is not enough to cover the heat loss from ambient heat exchange, at least outside tropical climates (Hagelqvist, 2009). Using anaerobic digestion can consume more energy than can be produced with the generated biogas (Hagelqvist, 2009; do Carmo Precci Lopes et al., 2018). The pulping processes often generate abundantly low temperature heat that can be used for this purpose with little or no operational cost. Sludge digestion is used before for example in Stora Enso mills in Heinola and Kotka in Finland (Ojanen, 2001). A novel case, Äänekoski mill, is discussed in more detail below.

#### 7.6.2 Case Äänekoski

Metsä Fibre Äänekoski mill is a modern pulp mill that started operation in 2017. The annual capacity of the mill is over 1 400 000 tons of air-dry pulp. Äänekoski mill utilizes side streams efficiently and produces several byproducts such as tall oil, sulphuric acid, and turpentine (Metsä Fibre, 2019a).

Äänekoski mill produces biogas from biosludge using anaerobic digestion process. Produced gas consists mainly of methane and carbon dioxide and is either burnt at the mill or refined further into more valuable products. The annual capacity of the anaerobic digestion plant is about 25 GWh of biogas and the yield is about 30% (Vakkilainen, 2018). The digestion process is not a final solution for disposal. Solid product from anaerobic digestion cannot be used for agricultural purposes due to low hygiene level. After digestion, sludge is dewatered mechanically and could be, for example, composted or used as fuel after suitable pretreatment. The volume of sludge after composting is about 30–40% of the volume before the digester (Metsä Fibre, 2014). Figure 7.13 shows the operating principle of the digestion system in Äänekoski.





#### 7.7 Land improvement

Sludges from pulp and paper mill processes can be used in land improvement and landscaping or as cover material for example in landfills. Pulp and paper sludge cannot

be awarded the European Union Community Ecolabel for soil improvers, because of the type of industry from which the sludge comes (EU 2015, criterion. 2.2). Composted sludge is, however, suitable for use in land improvement and landscaping as long as its heavy metal content does not exceed set limits (Ojanen, 2001). Sludge use in land improvement is a one of the ways to recirculate the nutrients, such as phosphorus and nitrogen, back to soil. Increasing use of forest residue and agricultural residue causes lack of nutrients in soil, which has several harmful effects for plants and the environment. Recycling of streams containing nutrients and organic substances is an essential part of circular bioeconomy, which is one of the hot topics today. In cultivation of food crops the limits for the content of harmful substances are stricter than in land improvement, and the use of sludges is also limited by general antipathy towards waste streams in connection with food.

Sludge use in land improvement always requires pretreatment, such as composting, decomposition, or lime stabilization. Composting is the most suitable pretreatment when sludge is used in landscaping or land improvement. Compared with municipal wastewater sludge, the sludges of forest industry contain less nutrients, but most likely also less pathogens. Sludge use in forests may harm the recreational use, but this can be reduced by pretreatment methods such as pelletizing or granulating. Composted sludge is typically mixed with ash before use for land improvement.

Table 7.2 gives the limits for the concentrations of harmful metals in sewage sludge used as a fertilizer fertilizers based on the EU Council Directive 86/278/EEC on soil protection when sewage sludge is used in agriculture and the limits in use in Finland. The Directive also covers other sludges than the urban sewage sludge and can therefore be applied to pulp mill sludge as well. The table also gives a comparison with the analysed pulp mill sludges presented in Appendixes A and B. The comparison shows that both the average and maximum contents are well below the limits in the analysed samples except for cadmium (Cd). Cd content was measured in all the samples presented in Appendix A and was below the allowed content in fertilizers only in one sample. High Cd content limits the use of sludge in areas not used for food production (Hagelqvist, 2013). Since the properties of sludges vary greatly, even the properties of different sludge streams in a single mill, analysis is needed before further use.

	Directive 86/278/EEC (EU, 1986)	Limits in Finland (Proagria, 2013)	Average content in samples	Maximum content in samples
As	-	25	3.9	5.6
Cd	20 to 40	1.5	5.7	8.6
Cr	-	300	44	60
Cu	1000 to 1500	$600^{1}$	35	42
Hg	16 to 25	1.0	0.4	0.6
Ni	300 to 400	100	40.6	70
Pb	750 to 1200	100	15.4	40
Zn	2500 to 4000	$1500^{1}$	489	920

Table 7.2. Limits for heavy metal content (mg/kgds) in sludge used as fertilizer compared with contents in sludge samples from Appendixes A and B.

<sup>1)</sup> Larger content is allowed in some cases when there is a lack of the given element in soil.

The current limits for harmful substances in soil production represent the situation today; it is likely that the limits are checked in the coming years. Therefore, the land use of sludges, either directly or after composting, cannot necessarily be considered a long-term solution (Norgren et al., 2015).

Sludges can be used in road structures depending on their properties, such as composition, water resistance, strength, and environmental safety. For example, the mixture of deinking sludge and fly ash has proved to be durable in road structures (Ojanen, 2001).

Clay is typically used as cover material in landfills. Especially paper mill sludges have similar sealing properties to clay. Fibre-rich sludge material is also less likely to crack than clay. The use of sludges decreases the covering costs and the use of non-renewable natural materials in landfills (Ojanen, 2001).

# 8 Effect of sludge disposal on energy consumption

The energy consumption of biosludge disposal processes affects the energy balance of the pulp mill. The effect can be estimated by calculating the mass and energy balances of the mill. Section 8.1 presents an estimation on how HTC treatment or anaerobic digestion of biosludge affects the energy balance of three different types of example mills. The implementation of the new processes is compared with traditional biosludge disposal processes in each mill case. The economic feasibility of the concepts is evaluated in section 0.

Traditionally, biosludge is incinerated in mill boilers, which affects the boiler operation and steam production. Sections 8.2 and 8.3 study the effect using a calculation template.

## 8.1 HTC and biogas processes in reference mills

#### 8.1.1 **Basis for the calculations**

The calculations are made based on the literature review, the results of laboratory-scale HTC trials conducted at LUT, and HTC process modelling. IPSEpro software was used to model the HTC unit. The HTC model is based on a previous study that evaluated HTC treatment of forest biomass (Saari et al., 2018). The pulp mill balances are calculated using an updated Millflow program that includes the detailed mass and energy balances of a kraft pulp mill. Millflow has been developed at LUT (Vakkilainen & Kivistö, 2008; Hamaguchi et al., 2011).

The Millflow balances are based on literature (Adams and Frederick, 1988; Grace and Malcolm, 1989; Gullichsen and Fogelholm, 1999a; Gullichsen and Fogelholm, 1999b), data from operating mills, and practical experience. Millflow has previously been used to pulp mill design by equipment vendors and in preliminary studies, for example by Vakkilainen and Kivistö (2008). The accuracy of the models has been verified in projects with pulp and paper companies and compared with other pulp mill models.

The Millflow programme includes detailed balances for each mill department (Hamaguchi et al., 2011). Figure 8.1 represents the primary inputs and outputs for each mill department. Table 8.1 gives the definitions for inputs and outputs presented in the figure. Pulp mill balances consist of hundreds of calculations, which makes it challenging to introduce them in detail. In the programme, the pulping line is dimensioned based on the desired capacity of the mill. The wood demand is calculated based on the cooking yield and losses in the process stages, and the chemicals demand is calculated from the digester balance. The programme includes a black liquor calculation tool to define the inputs for the recovery boiler calculations, such as the dry solids flow, the composition, and the heating value of black liquor. The black liquor calculation tool is partly based on correlations from a black liquor database (Vakkilainen, 2000), and Hamaguchi et al. (2011) improved it by taking into account the lignin removal. The lime kiln calculations require that inputs such as the type of make-up lime, the amount of residual lime, and the availability of burnt lime be defined.

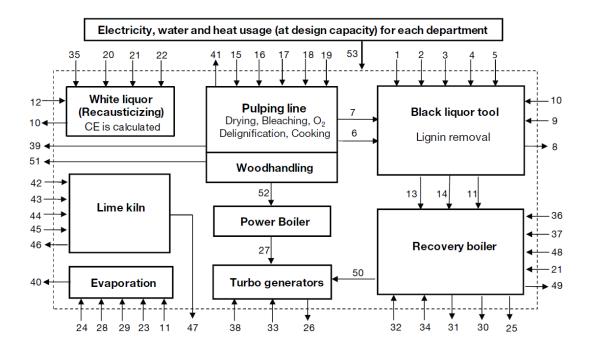


Figure 8.1. The main inputs and outputs for the Millflow balances. (Hamaguchi et al., 2011)

The electricity generation is calculated from the turbogenerator balances after defining the steam flows from the recovery boiler and the biomass boiler. The electricity and heat usage per unit of production in each department of the mill is used to define the steam balance. The condensing tail flow sets the steam balance. Literature values are used to define typical departmental energy consumption (Nieminen, 2007; Vakkilainen & Kivistö, 2014). During this study, an HTC unit based on the modelling was added to calculate its effect on the balance. The biogas cases are calculated based on the assumption that the produced gas and digestate do not return to the mill process after digestion.

Table 8.1. Description of inputs and outputs presented in Figure 8.1. Modified from Hamaguchi et al. (2011).

ID	Description	ID Description
1	Lignin heating value, MJ/kg	27 HP steam flow, power boiler, t/h
2	Wood composition, wt-%	28 Strong liquor virgin concentration, %
3	Heating values of inorganics (Na <sub>2</sub> S, K <sub>2</sub> S)	29 Steam economy, kJ/kgH <sub>2</sub> O
4	Organics heating values (acids, etc)	30 Flue gas generation, Nm <sup>3</sup> /h
5	Washing efficiency of pulp, %	31 Air ratio, excess air, %
6	Dry wood demand, t/h	32 Blowdown and sootblowing, kg/s
7	Dry unbleached pulp, t/d	33 All steam requirements (kg/s, °C, bar)
8	Lignin in pulp, kg/ADt	34 HP steam parameters (°C, bar)
9	Lignin removal rate, %	35 Cl and K content in liquor, wt-%
10	Inorganic compounds in WL, kg/BDt	36 Combustion air temperatures, °C
11	Dry solids generation tDS/d	37 Recycled ash (% of variable 11)
12	Formation rate of $S_2O_3^{2-}$ and $SO_3^{2-}$ , %	38 Condensate return from the departments, t/h
13	Black liquor heating value, MJ/kg	39 Recausticizing required capacity, m <sup>3</sup> WL/d
14	Black liquor composition, wt-%	40 Evaporation required capacity, t/h
15	Cooking yield, %	41 Pulping line required capacity, ADt/d
16	Equipment losses, %	42 Availability of burnt lime, %
17	Effective alkali charge as NaOH, %	43 Availability of make-up lime, %
18	Desired bleached pulp capacity, ADt/a	44 Residual lime and CaCO <sub>3</sub> in lime mud, %
19	Dilution water in pulp washing, kg/ADt	45 Kiln heat requirement, MJ/kg
20	Sulfidity, %	46 Amount of lignin to be burned, t/d
21	Reduction, %	47 Kiln required capacity, t lime/d
22	Active alkali charge, gNaOH/l	48 Recovery boiler furnace area, m <sup>2</sup>
23	MP steam/total steam usage, %	49 HHRR and heat load, kW/m <sup>2</sup> and MW
24	Additional load to be evaporated, t/h	50 HP steam flow, recovery boiler, t/h
25	Smelt production, kg/s	51 Wood handling required capacity, m <sup>3</sup> /d
26	Power generation, MW	52 Bark to burning, BDt/d

The studied mills represent typical mills in their chosen location. Two mills are located in Scandinavia (Mills NorPulp and NorInt) and one in South America (Mill Euca). The main operational parameters of the mills are collected in Table 8.2. For the northern mills, the HTC cases are calculated using two different amounts of biosludge.

	Unit	Euca	NorPulp	NorInt
Production			-	
Operating hours	h/a	8400	8400	8400
Pulp production	ADt/d	4286	1714	4000
Paper production	t/d	-	-	4224
Wood handling				
Wood income	BDt/d	8405	4390	10 007
Wood type		eucalyptus	softwood	softwood
Residue	BDt/d	638	753	1541
Wood moisture	-	45%	50%	50%
Recovery boiler				
Dry solids to boiler	tds/d	6783	3445 / 3454	7997
Net steam flow	t/h	873	444 / 445	1050
Biomass boiler				
Biomass fuel use	BDt/d	-	-	1514
Net steam flow	t/h	-	-	309 / 308
Biosludge				
Production	BDkg/ADt	11	5 / 10	5 / 10
Production	tds/d	48	8.6 / 17.1	20 / 40
Energy				
Steam use, pulp mill	t/h	662	343 / 344	781
Steam use, paper mill	t/h	-	-	401
Power generation	MW	160	72	233 / 232
Power consumption, pulp mill	kWh/ADt	501	59	552
Power consumption, paper mill	kWh/t	-	-	681

Table 8.2. The reference mill operations and the main process flows. For the northern mills, two separate cases are considered using biosludge production of 5 and 10 BDkg/ADt.

The southern mill (Mill Euca) is a large, modern stand-alone kraft pulp mill producing bleached pulp from eucalyptus. Its annual pulp production is 1.5 million ADt. The sole energy source is the recovery boiler, and no separate power boiler exists. The recovery boiler is able to cover the energy demand of the mill, which is typical for a modern stand-alone mill. Biosludge is dried mechanically and mixed with weak black liquor at the evaporation stage. It is then combusted in the recovery boiler. Biosludge production is 11.2 BDkg/ADt. Steam from the recovery boiler is used in the mill processes and for

power generation in a turbogenerator. Low temperature heat flows are abundantly available to, for example, drying purposes. The electricity generation exceeds the mill power requirements, and thus the sale of excess electricity is possible. Hamaguchi et al. (2011) presented a more detailed description on the design conditions of the mill, and a detailed balance can be found in (Kuparinen, 2019).

The studied northern stand-alone mill (Mill NorPulp) produces 600 000 ADt/a bleached pulp. The mill uses softwood as raw material. Steam is produced in the recovery boiler and there is no separate power boiler. Mechanically dried biosludge is mixed with black liquor before evaporation and incineration in the recovery boiler. Steam is used firstly in the mill processes and secondly, in a turbogenerator for electricity generation. The mill is self-sufficient in energy and able to sell excess electricity.

The third reference mill is a large northern integrated pulp and paper mill (Mill NorInt). The mill produces bleached pulp from softwood and uses its production for papermaking. Its annual pulp production is 1.4 million ADt. The integrated paper mill produces coated and uncoated paper and is a notable additional energy consumer. The recovery boiler and a separate biomass boiler produce steam for use in pulp and paper production and for electricity generation in a turbogenerator. All residue from the wood handling process is combusted in the biomass boiler. Biosludge is dried mechanically and combusted with wood residue in the biomass boiler. Kuparinen (2019) presented more detailed mass and energy balances for the mill.

Biosludge properties vary depending on process details. In these calculations, typical composition based on earlier data (see section 3.1 and Appendixes A and B) was assumed. The composition of biosludge affect its treatment processes. Regarding the combustion processes, the effect of moisture content notably exceeds the effect of biosludge composition. Therefore, further analysis on the composition is not needed for the HTC process. Four separate HTC cases were calculated for each reference mill as follows:

- **Case 0**: Conventional biosludge disposal by incineration either in the recovery boiler or in the power boiler after mechanical drying/drying with black liquor in the evaporators.
- **Base**: HTC of biosludge with mechanical drying (filtration) before and after the process followed by incineration of hydrochar in either the recovery boiler or the power boiler. Hydrochar dry matter content before incineration 16.5%.
- **OptA**: HTC of biosludge with mechanical drying (filtration) before and after the process followed by incineration of hydrochar in either the recovery boiler or the power boiler. Heat recovery from the HTC process used for biosludge preheating in a heat exchanger. Hydrochar dry matter content before incineration 35%.
- **OptB**: HTC of biosludge using mechanical drying (filtration) before and after the process followed by incineration of hydrochar in either the recovery boiler or the power boiler. Heat recovery from the HTC process to preheat the incoming biosludge and for thermal drying of hydrochar. Hydrochar dry matter content before incineration 90%.

In all the cases, biosludge is dried mechanically before further treatment. The scenarios assume that there is existing equipment for drying. Moisture content of biosludge is estimated at 92% after drying. For raw biosludge, a higher heating value of 21 MJ/kgds was estimated based on recent sludge analysis at LUT. In the cases where biosludge is incinerated in the recovery boiler, the capacity of evaporators is kept constant. When HTC is implemented and biosludge is no longer conveyed to the evaporators, this leads to the possibility to increase the dry matter content of black liquor. The wastewater from the HTC process is led to the existing water treatment system at the mills in all the cases.

Hydrothermal carbonization of biomass typically produces hydrochar that is hydrophobic and can easily be dried into a high dry matter content. Properties of hydrochar can change the situation; biosludge drying is exceptionally difficult due to strong cell walls that prevent the removal of intracellular water. It is difficult to estimate based on the lab-scale trials whether the HTC process is capable of breaking the cell walls. However, it can be assumed that in an industrial HTC process where hydrochar slurry is transferred from the reactor to the flash tanks, the rapid pressure changes break the cell walls. The following assumptions were made for the HTC model:

- Dry matter content of biosludge after mechanical drying 8%
- Biosludge temperature 10 °C and pressure 1 bar before HTC
- Residence time 3 h
- Reactor temperature 200 °C
- Hydrochar yield 75% of dry matter
- Hydrochar LHV 23.94 MJ/kgds
- Electricity consumption in HTC process
  - Mechanical drying of hydrochar 150 kW for 1.3 kgds/s
  - Feed pumps <150 kW
  - Thermal drying of hydrochar 300 kW for 1.3 kgds/s (only in OptB)

Anaerobic digestion (AD) process is used to produce biogas from biosludge. The process occurs in the absence of oxygen at 30-38 °C. Organic material decomposes forming biogas and the residue forms digestate. The formed biogas consists mainly of methane and carbon dioxide. Biogas can be used in combustion processes. In a pulp mill, it can for example partly substitute for fossil fuels in the lime kiln. The heating value of biogas is around 19–23 MJ/m<sup>3</sup>. The yield and properties of the produced biogas depend greatly on sludge properties and process details. In these calculations the end-use of biogas has been left open and does not affect the mill balances. The calculations focus on the changes in the energy balance when biosludge is no longer combusted in the mill boilers. Electricity use for the digestion process is accounted and assumed at 0.2 MWh per ton of untreated dried sludge based on the results of Stoica et al. (2009). Low temperature heat from the mill processes is used for heating the digestion reactors. The effect on the energy balance is considered negligible due to abundant low temperature heat steams in the process. Further analysis on the use of biogas would require more detailed modelling of the digestion process. In all the cases, the total biosludge stream is led to the digestion process.

#### 8.1.2 Southern pulp mill case – Euca

In the southern reference mill, biosludge production is 11.2 BDkg/ADt, corresponding to 48 tds/d. Case 0 represents a case where biosludge is incinerated with black liquor in the recovery boiler after mechanical drying and the evaporation stage. Figure 8.2 shows the

simplified block diagram of the Euca mill in case 0. Biosludge flow includes significant amount of water that needs to be evaporated thus decreasing the capacity that can be used for black liquor evaporation.

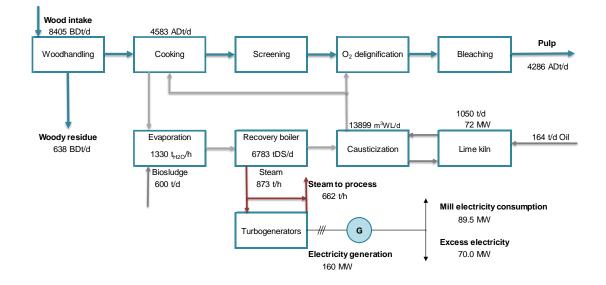


Figure 8.2. The flow diagram of the Euca mill process in Case 0.

When the HTC process is integrated in the mill process, produced hydrochar is led to the recovery boiler. Figure 8.3 shows the main process flows in the Euca mill in HTC Base case, where HTC-treated biosludge is combusted in the recovery boiler at 16.5% dry matter content. When the evaporation capacity is kept constant, black liquor dry matter content can be increased from 79% at Case 0 to 85% at Base case. Considering medium pressure (MP) steam use for HTC, the net increase in steam production compared with Case 0 is 15 t/h. The main reason for the increase is the higher moisture content of black liquor, and the improved combustion properties of biosludge due to HTC treatment have minor impact. Consequently, electricity production capacity increases by 4.85 MW due to increased steam production, considering also the steam and electricity used for HTC.

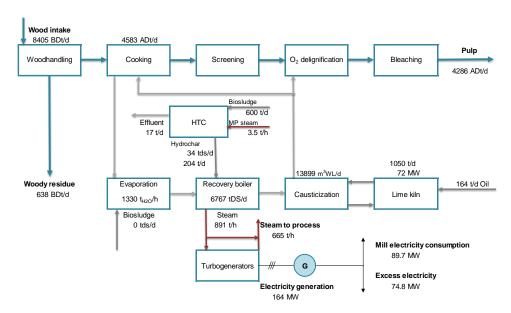


Figure 8.3. The flow diagram of the Euca mill process in the HTC Base case.

In case OptA the energy efficiency of the HTC process in improved by using recovered heat to preheat the biosludge flow before the HTC reactor. Electricity production slightly increases compared with the Base case. The steam use is lower in OptA case than in the Base case due to heat recovery. The hydrochar dry matter content increases from 16.5% to 35%, which has a small impact on steam production in the recovery boiler. Figure 8.4 shows the main process flows in the OptA case.

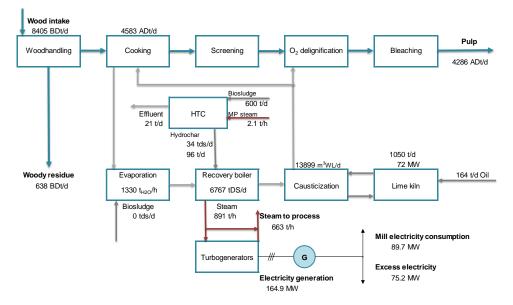


Figure 8.4. OptA case of biosludge HTC treatment implemented in the Euca mill.

In the Opt B case, thermal drying using recovered heat from the HTC process is added to increase the dry matter content of hydrochar up to 90%, given that HTC breaks the cell walls and enables the removal of intercellular water. Figure 8.5 shows the block diagram of OptB case implementation in the Euca mill. The thermal drying process doubles the electricity consumption compared with OptA case, and also makes the process more complicated thus increasing costs. However, the increased dry matter content of hydrochar only has a small impact on steam production in the recovery boiler. Considering higher steam and electricity consumption compared with OptA, OptB does not seem feasible since the amount of sellable electricity is lower than in OptA.

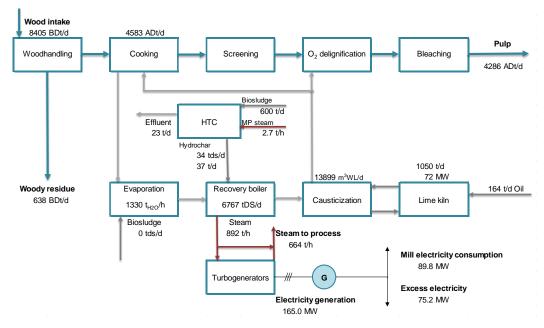


Figure 8.5. OptB case of biosludge HTC treatment implemented in the Euca mill.

The main benefit of the HTC treatment in the Euca mill is the possibility to increase steam production followed by increase in electricity generation. When electricity is a sellable product, the net change in the electricity balance is the primary parameter when feasibility is evaluated. The main reason for additional steam production is the possibility to use the evaporation capacity to evaporate the black liquor flow into higher moisture content, when the biosludge flow including significant amount of water is no longer led to the evaporators. HTC process also increases the heating value of biosludge dry solids and enables biosludge drying into higher moisture content due changes in biosludge properties. In all the HTC cases, net change in electricity generation is positive compared with the 0-case (Table 8.3). OptA can be considered most feasible of these cases due to highest increase in sellable electricity. The thermal drying of hydrochar in OptB case does not bring additional benefit since the increased energy production does not cover the increased consumption compared with OptA.

	Unit	0-case	Base	OptA	OptB
Biosludge/hydrochar dry matter	%	8.0 %	16.5 %	35 %	90 %
Steam use in HTC	t/h	-	3.48	2.06	2.68
Electricity use in HTC	MW	-	0.14	0.14	0.28
Steam production in recovery boiler	t/h	873	891	891	892
Net change in steam <sup>1</sup>	t/h	-	15.08	16.70	16.90
Electricity generation	MW	160	164	165	165
Net change in electricity <sup>1</sup>	MW	-	4.85	5.27	5.23

Table 8.3. Comparison of the energy balances for the cases on the HTC treatment of biosludge in the southern reference mill (Euca).

<sup>1</sup>Compared with 0-case

Anaerobic digestion of biosludge changes the mill operations because wet sludge is no longer led to evaporation and further to the recovery boiler. When the evaporation capacity is kept constant similarly as in the HTC cases, higher black liquor dry matter content is achieved in the evaporators without additional load of the biosludge. This leads to increased steam production in the recovery boiler. The steam and electricity consumption of the boiler slightly decreases due to smaller load in the boiler, but electricity consumption increases due to consumption in AD. Table 8.4 collects the main changes in the energy balance of the mill in the AD case. Figure 8.6 shows the main balance for the mill in the AD case.

	Unit	0-case	AD case
Change in steam consumption <sup>1</sup>	t/h	-	-0.52
Steam production in recovery boiler	t/h	873	878
Net change in steam <sup>1</sup>	t/h	-	6.16
Electricity consumption in AD	MW	-	0.4
Electricity generation	MW	160	161
Net change in electricity <sup>1</sup>	MW	-	1.57

Table 8.4. Effect of anaerobic digestion of biosludge on the energy balance in the southern reference mill case.

<sup>1</sup>Compared with 0-case

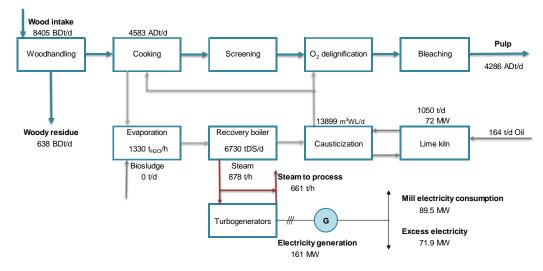


Figure 8.6. Euca mill balance when biosludge is treated separately in the anaerobic digestion process.

In a pulp mill where the recovery boiler is the bottleneck of the pulp production and biosludge is disposed of by incineration in the recovery boiler, HTC or AD processes can offer a novel way to increase the pulp production capacity. Hydrochar incineration instead of untreated biosludge decreases the load in the recovery boiler, which enables increased amount of black liquor in the boiler. The AD case leads similarly to decreased load in the recovery boiler since biosludge is handled elsewhere.

#### 8.1.3 Northern stand-alone pulp mill – NorPulp

To gain a more comprehensive view on the effect of the HTC of biosludge on the mill balances, the process is estimated for the northern mill cases using two different biosludge amounts. In pulp mills, the amount of biosludge depends on the process details and varies between mills. It can be assumed that the changes in energy balances are clearer for mills with larger amount of biosludge.

NorPulp1 represents the northern stand-alone mill case where biosludge production is 5 BDkg/ADt corresponding to 8.6 tds/d. Figure 8.7 shows the main process flows of NorPulp1 in 0-case, where biosludge is led to the evaporators and further to the recovery boiler for combustion with black liquor.

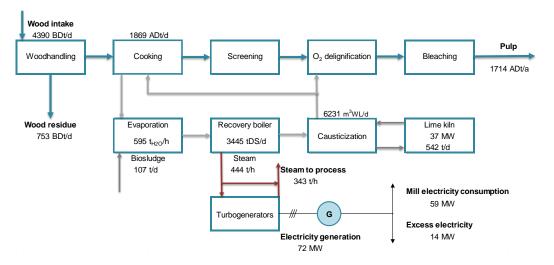


Figure 8.7. The main process flows of NorPulp1 mill for Case 0.

In the HTC base case (Figure 8.8), a small increase in steam production followed by increase of electricity generation can be detected compared with the 0-case. Similarly as for the Euca mill, higher black liquor dry solids content can be achieved when biosludge is treated with HTC instead of conveying to the evaporators, since the evaporation capacity remains unchanged.

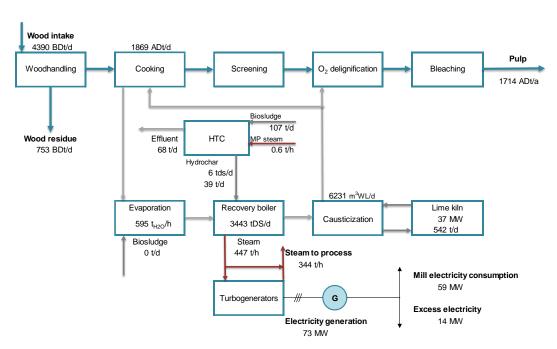


Figure 8.8. The flow diagram of the NorPulp1 mill process in the HTC base case.

The OptA case for the mill NorPulp1 is depicted in Figure 8.9. Increased energy efficiency via heat recovery and biosludge preheating leads to lower moisture content of produced hydrochar. Also, less steam is needed than in the 0-case.

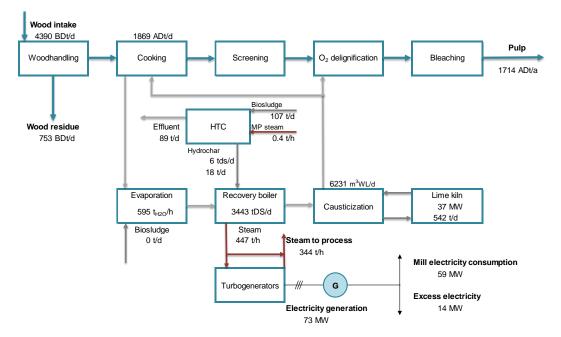


Figure 8.9. The OptA case of the HTC treatment implemented in NorPulp1 mill.

The OptB case includes the thermal drying of hydrochar resulting in 90% dry matter content before combustion in the recovery boiler. Similarly as for the Euca mill, the energy production increase due to increased hydrochar dry matter content does not cover the energy consumption of the thermal drying. Therefore, slightly less electricity is available for sale than in OptA case. The process flows of the OptB case for NorPulp1 are presented in Figure 8.10.

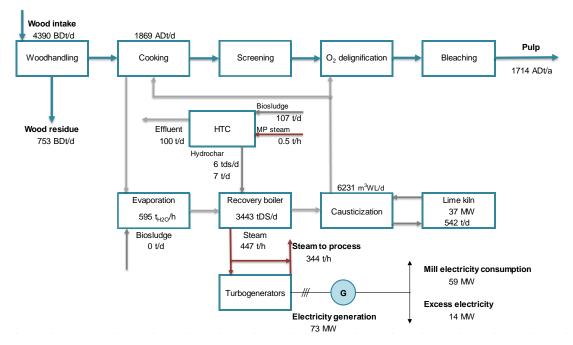


Figure 8.10. The process flows of NorPulp1 mill in the OptB case.

NorPulp2 represents the same stand-alone mill case with biosludge amount of 10 BDkg/ADt corresponding to 17.1 tds/d. Figure 8.11 shows the main process flows of NorPulp2 in 0-case, where biosludge is mixed with black liquor before evaporators and incinerated in the recovery boiler. A higher evaporation capacity is needed compared with NorPulp1 due to larger amount of biosludge in the process.

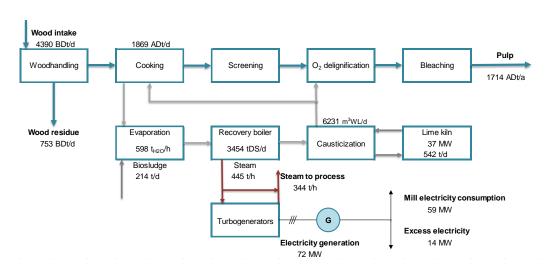


Figure 8.11. The main process flows of NorPulp2 mill for Case 0.

The HTC base case for NorPulp2 mill is presented in Figure 8.12. A clearer energy production increase can be seen compared with NorPulp1 due to higher amount of biosludge.

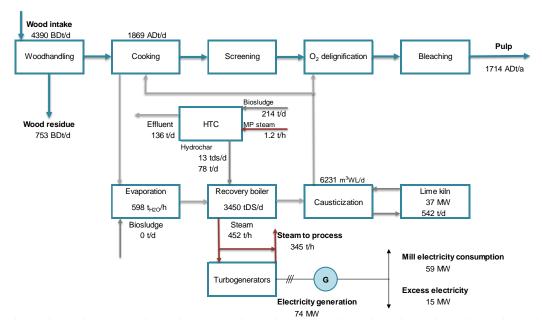


Figure 8.12. The flow diagram of NorPulp2 mill in the HTC base case.

In the OptA case, where recovered heat is used to preheat the biosludge flow before HTC reactor, a small increase in electricity production can be seen compared with the base case. Figure 8.13 shows the main process flows for NorPulp2 mill in the OptA case.

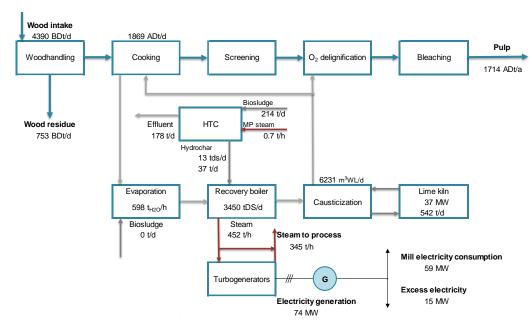


Figure 8.13. The main process flows for NorPulp2 mill in the OptA case.

The OptB case (Figure 8.14) shows how the NorPulp2 mill balances change when the thermal drying of hydrochar is added.

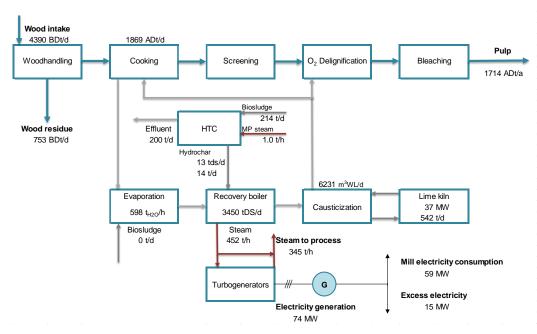


Figure 8.14. The main process flows for NorPulp2 mill in the OptB case.

Table 8.5 compares the HTC cases in the mills NorPulp1 and NorPulp2. Similarly as in the Euca mill cases, the main direct benefit of the HTC treatment is the possibility to

increase electricity generation via increased steam production. The heating value of untreated biosludge is very low, in some cases negative (see Figure 3.1.). Steam production can be increased, and more electricity produced in all the HTC cases compared with the 0-case, mainly because less energy is used to evaporate water included in biosludge in the furnace. As could be assumed, the change is clearer when the amount of biosludge is larger. As for the Euca mill, the main difference between the 0-case and the HTC cases is the possibility to increase the dry solids content of black liquor due to available capacity at the evaporation stage. The increase in the heating value and dry matter content of hydrochar explains the differences between the HTC cases Base, OptA, and OptB. The net change in electricity is the highest in OptA case. In OptB, the steam production increase due to lower moisture content of hydrochar does not cover the increased energy consumption in HTC process.

NorPulp1, biosludge 5 BDkg/ADt	Unit	0-case	Base	OptA	OptB
Biosludge/hydrochar dry matter	%	8.0%	16.5%	35%	90%
Steam use in HTC	t/h	-	0.62	0.37	0.48
Electricity use in HTC	MW	-	0.02	0.02	0.05
Steam production in recovery boiler	t/h	444	447	447	447
Net change in steam <sup>1</sup>	t/h	-	2.56	2.89	2.94
Electricity generation	MW	72.1	72.9	73.0	73.0
Net change in electricity <sup>1</sup>	MW	-	0.76	0.84	0.83
NorPulp2, biosludge 10 BDkg/ADt	Unit	0-case	Base	OptA	OptB
Biosludge/hydrochar dry matter	%	8.0%	16.5%	35%	90%
Steam use in HTC	t/h	-	1.24	0.74	0.96
Electricity use in HTC	MW	-	0.05	0.05	0.10
Steam production in recovery boiler	t/h	445	452	452	452
Net change in steam <sup>1</sup>	t/h	-	5.39	6.00	6.07
Electricity generation	MW	72.4	74.0	74.1	74.2
Net change in electricity <sup>1</sup>	MW	_	1.58	1.74	1.72

Table 8.5. The comparison of the energy balances for the biosludge treatment cases in the northern stand-alone reference mill (NorPulp).

<sup>1</sup>Compared with 0-case

Anaerobic digestion of biosludge decreases the load of the recovery boiler and evaporators when biosludge is handled separately. Assuming the constant evaporation capacity similarly as in the HTC cases, higher black liquor dry solids content is achieved in the evaporators in the AD cases. This leads to increase in recovery boiler steam production and consequently to higher electricity generation. Figure 8.15 shows the main flows for NorPulp1 and Figure 8.16 for NorPulp2 in the AD case. The differences between the cases result from higher evaporation capacity in NorPulp2 because it was scaled based on a higher amount of biosludge.

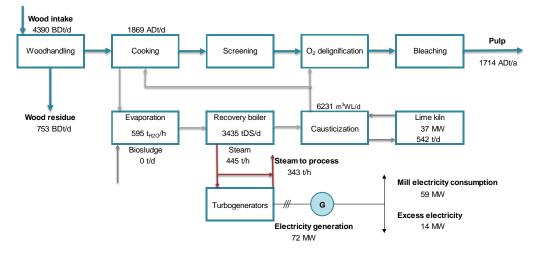


Figure 8.15. NorPulp1 mill balance in the AD case.

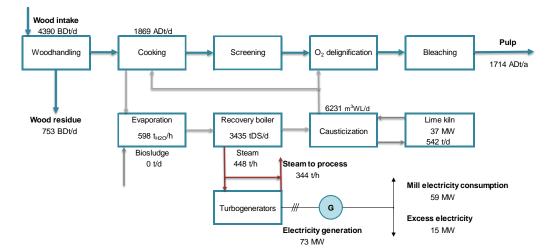


Figure 8.16. NorPulp2 mill balance in the AD case.

Table 8.6 presents the changes in steam and electricity balances due to AD process compared with the 0-case. Minor difference in steam and electricity consumption in the recovery boiler operations can be detected. The main difference is the higher steam and electricity production, but part of the produced additional electricity is consumed in AD.

NorPulp1, biosludge 5 BDkg/ADt	Unit	0-case	AD case
Change in steam consumption <sup>1</sup>	t/h	-	-0.06
Steam production in recovery boiler	t/h	444	445
Net change in steam <sup>1</sup>	t/h	-	1.24
Electricity consumption in AD	MW	-	0.07
Electricity generation	MW	72.1	72.4
Net change in electricity <sup>1</sup>	MW	-	0.29
NorPulp2, biosludge 10 BDkg/ADt	Unit	0-case	AD case
Change in steam consumption <sup>1</sup>	t/h	-	-0.08
Steam production in recovery boiler	t/h	445	448
Net change in steam <sup>1</sup>	t/h	-	3.02
Electricity consumption in AD	MW	-	0.14
Electricity generation	MW	72.4	73.2
Net change in electricity <sup>1</sup>	MW	_	0.74

Table 8.6. Effect of anaerobic digestion of biosludge on the energy balance in the NorPulp mill cases.

<sup>1</sup>Compared with 0-case

# 8.1.4 The integrated pulp and paper mill case – NorInt

The energy balances for the northern integrated pulp and paper mill (NorInt) were calculated for cases with two biosludge amounts. NorInt1 represents a case where biosludge production is 5 BDkg/ADt, corresponding to 20 tds/d. Figure 8.17 shows the main process flows of NorInt1 in 0-case, where biosludge is incinerated with biomass residue in the biomass boiler. The moisture content of biomass before combustion is 92%.

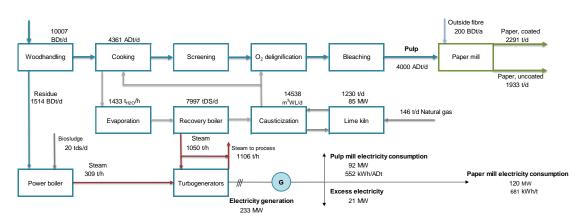


Figure 8.17. The block diagram of NorInt1 mill in 0-case, where mechanically dried biosludge is incinerated in biomass boiler with biomass residue.

In the HTC Base case biosludge is HTC-treated in the simple process with only mechanical drying of the produced hydrochar. Steam production in the boiler increases due to less water in the combustion process and due to slightly higher heating value of the hydrochar dry solids compared with biosludge. The case is depicted in Figure 8.18.

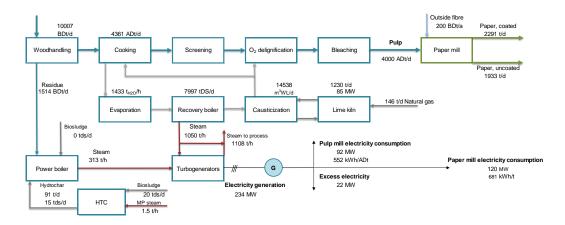


Figure 8.18. The process flows of the HTC base case for the NorInt1 mill.

In the OptA case heat recovery is used to preheat the biosludge flow before HTC reactor. Consequently, less steam is needed but higher hydrochar ds content (35%) is achieved. The process flows for the NorInt1 mill in case OptA are presented in Figure 8.19.

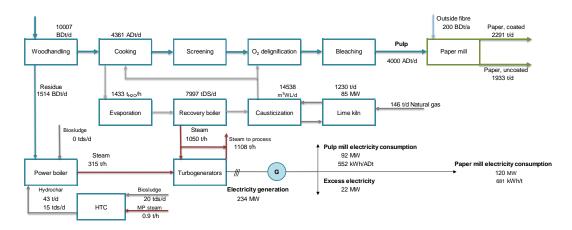


Figure 8.19. The process flows of the OptA case for the NorInt1 mill.

In the OptB case, 90% dry matter content of biosludge is achieved by adding thermal drying of hydrochar. Figure 8.20 presents the case for NorInt1 mill.

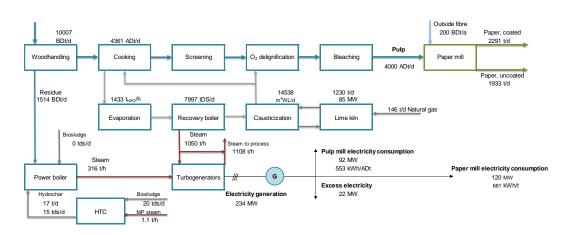


Figure 8.20. The process flows of the OptB case for the NorInt1 mill.

NorInt2 represents the integrated mill case where biosludge production is 10 BDkg/ADt corresponding to 40 tds/d. Figure 8.21 shows the main process flows of 0-case without HTC process, when biosludge is incinerated with biomass residue in the biomass boiler at 8% dry matter content.

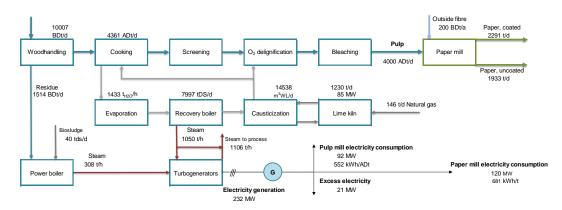


Figure 8.21. The main process flows of the NorInt2 mill in the 0-case.

Figure 8.22 shows the main process flows of the HTC Base case for the NorInt2 mill. The figure shows that steam production increases due to less water in the biomass boiler during combustion. Figure 8.23 presents the OPtA case for the mill NorInt2. Figure 8.24 shows the main process flows of OptB case for the NorInt2 mill.

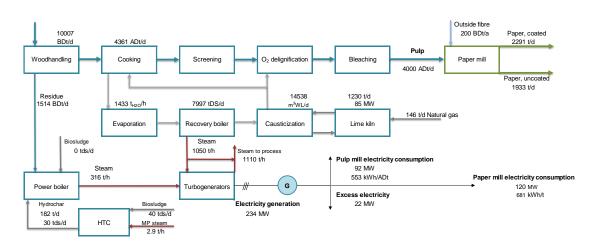


Figure 8.22. The block diagram of the NorInt2 mill the HTC base case.

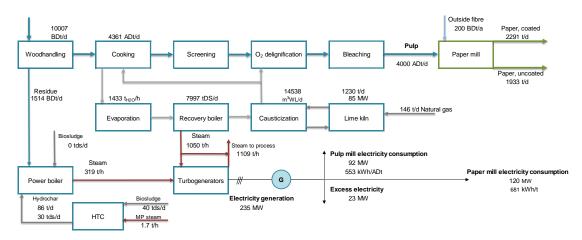


Figure 8.23. The process flows of the OptA case for the NorInt2 mill.

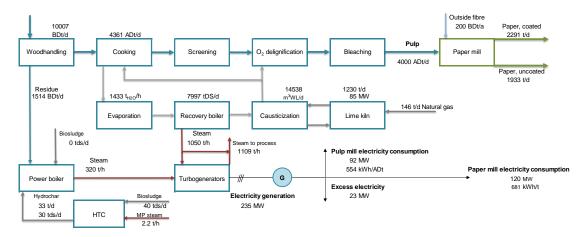


Figure 8.24. The process flows of the OptB case for the NorInt2 mill.

Table 8.7 compares the energy balances for the HTC cases in the NorInt mill. For this mill case, OptB gives the highest net change in electricity unlike in the Euca and NorPulp mills. It is, however, unlikely that investing in the relatively expensive thermal drying equipment in OptB would be economically feasible when the additional electricity compared with OptA is only 0.1 and 0.2 MW for the mills NorInt1 and NorInt2, respectively. Therefore, it is probable that as for the Euca and NorPulp mills, OptA is the most feasible option also for this mill.

When biosludge is treated with anaerobic digestion instead of incineration in the biomass boiler, the amount of water into the biomass boiler decreases. In the example cases, this leads to increase in steam production. Figure 8.25 presents the main mill flows for the NorInt mill when biosludge is handled separately using AD. When the biosludge is no longer incinerated in the biomass boiler, the clear differences between the energy balances of NorInt1 and NorInt2 cases disappear.

NorInt1, biosludge 5 BDkg/ADt	Unit	0-case	Base	OptA	OptB
Biosludge/hydrochar dry matter	%	8.0%	16.5%	35%	90%
Steam use in HTC	t/h	-	1.45	0.86	1.12
Electricity use in HTC	MW	-	0.06	0.06	0.12
Steam production in biomass boiler	t/h	309	313	315	316
Net change in steam <sup>1</sup>	t/h	-	2.57	4.65	5.21
Electricity generation	MW	233	234	234	234
Net change in electricity <sup>1</sup>	MW	-	0.72	1.26	1.37
NorInt2, biosludge 10 BDkg/ADt	Unit	0-case	Base	OptA	OptB
Biosludge/hydrochar dry matter	%	8.0%	16.5%	35%	90%
Steam use in HTC	t/h	-	2.90	1.72	2.23
Electricity use in HTC	MW	-	0.12	0.12	0.23
Steam production in biomass boiler	t/h	308	316	319	320
Net change in steam <sup>1</sup>	t/h	-	5.14	9.31	10.42
Electricity generation	MW	232	234	235	235
Net change in electricity <sup>1</sup>	MW	-	1.43	2.52	2.73

Table 8.7. The comparison of the energy balances for the biosludge HTC treatment cases in the northern integrated reference mill (NorInt).

<sup>1</sup>Compared with 0-case

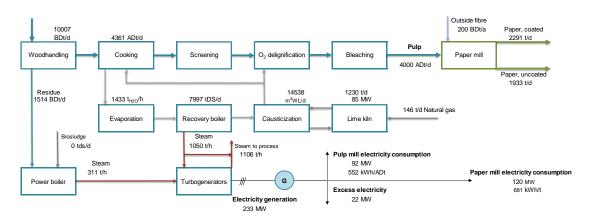


Figure 8.25. NorInt reference mill flows when biosludge is handled separately using anaerobic digestion.

Table 8.8 presents the comparison of the energy balances between 0-case and AD case in the NorInt1 and NorInt2 mills. The negative effect of incinerating wet biosludge in the biomass boiler can be seen as increased steam and electricity production when biosludge flow is led to a separate process. A small increase in the steam and power consumption of the biomass boiler can be seen due to the change.

NorInt1, biosludge 5 BDkg/ADt	Unit	0-case	AD case
Change in steam consumption <sup>1</sup>	t/h	-	0.11
Steam production in biomass boiler	t/h	309	311
Net change in steam <sup>1</sup>	t/h	-	1.69
Electricity consumption in AD	MW	-	0.17
Electricity generation	MW	233	233
Net change in electricity <sup>1</sup>	MW	-	0.33
NorInt2, biosludge 10 BDkg/ADt	Unit	0-case	AD case
Change in steam consumption <sup>1</sup>	t/h	-	0.22
Steam production in biomass boiler	t/h	308	311
Net change in steam <sup>1</sup>	t/h	-	3.37
Electricity consumption in AD	MW	-	0.33
Electricity generation	MW	232	233
Net change in electricity <sup>1</sup>	MW		0.65

Table 8.8. The comparison of the energy balances for the AD cases in the NorInt mill.

<sup>1</sup>Compared with 0-case

### 8.1.5 Discussion on the case studies

Sludge disposal method affects the energy balance of the mill. Biosludge disposal was taken in the focus of the estimation of the new processes since biosludge is more difficult to dispose of then primary sludge, and the studied processes were considered suitable for biosludge disposal. HTC and AD can be used to refine biosludge into a usable form, and the results show the effect of these processes on the mill energy balance in comparison with conventional methods to dispose of biosludge by incineration in the mill boilers. Primary sludge has more usage possibilities and its conversion for energy products is therefore not the primary option considering for example the waste management hierarchy presented in Figure 7.1.

Table 8.9 presents a summary of the cases studied above using the reference mill models. Biosludge properties vary greatly depending e.g. on the process and mill practices. Recent biosludge analysis results from LUT were used here as a starting point for modelling. The results show, that even though biosludge is a small portion of the fuel flow in the boilers, alternative handling methods can improve the boiler operation and consequently increase steam and electricity consumption. The economic feasibility of the studied concepts is also an important question, and it is further discussed in section 0.

In the HTC cases produced hydrochar is used in the mill for energy, which shows in the results. Hydrochar use for other purposes, such as soil conditioning, would slightly lower the amount of produced heat and power generated but might offer other benefits, for example in the form of sellable products, such as activated carbon. A future possibility would be to utilize hydrochar as carbon storage if the mill would benefit from negative carbon dioxide emissions.

	Unit	Euca	NorPulp	NorInt
Operation				
Operating hours	h/a	8400	8400	8400
Pulp production	ADt/d	4286	1714	4000
Paper production	t/d	-	-	4224
Wood type		eucalyptus	softwood	softwood
Biosludge				
Production	BDkg/ADt	11	5 / 10	5 / 10
Production	tds/d	48	8.6 / 17.1	20 / 40
Disposal in base case		RB	RB	BB
Dry matter after filtration	%	8%	8%	8%
Heating value (HHV)	MJ/kgds	21	21	21
Energy, 0-case	U			
Steam use, pulp mill	t/h	662	343 / 344	781
Steam use, pulp mill	t/h	662	343 / 344	781
Steam use, paper mill	t/h	-	-	401
Power generation	MW	160	72	233 / 232
Power consumption, pulp mill	kWh/ADt	501	59	552
HTC				
Biochar heating value (LHV)	MJ/kgds	24	24	24
Biochar use	-	RB	RB	BB
Base case				
Hydrochar dry matter content	%	16.5%	16.5%	16.5%
Net change in electricity <sup>1</sup>	MW	4.85	0.67 / 1.58	0.72 / 1.43
Net change in steam <sup>1</sup>	t/h	15.08	2.56 / 5.39	2.57 / 5.14
Opt A case				
Hydrochar dry matter content	%	35%	35%	35%
Net change in electricity <sup>1</sup>	MW	5.27	0.84 / 1.74	1.26 / 2.52
Net change in steam <sup>1</sup>	t/h	16.70	2.89 / 6.00	4.65 / 9.3
Opt B case				
Hydrochar dry matter content	%	90%	90%	90%
Net change in electricity <sup>1</sup>	MW	5.23	0.83 / 1.72	1.37 / 2.73
Net change in steam <sup>1</sup>	t/h	16.90	2.94 / 6.07	5.21 / 10.4
AD				
Net change in electricity <sup>1</sup>	MW	1.57	0.29 / 0.74	0.33 / 0.63
Net change in steam <sup>1</sup>	t/h	6.16	1.24 / 3.02	1.69 / 3.3
Compared with 0-case				

Table 8.9. HTC and biogas cases for the reference mills. RB refers to recovery boiler and BB to biomass boiler

<sup>1</sup>Compared with 0-case

The AD cases do not consider the use of produced biogas, but it could be utilized in the mill, for example in the lime kiln. Although, refining further for a sellable product can be a more feasible option, for example when there is demand for biogas for vehicle use in the neighbourhood. The removal of wet biosludge from the process seems to have a positive effect on mill energy balances regardless of the end use of sludge product.

Incineration of wet sludge is primarily a disposal method; it cannot be justified energywise.

## 8.2 Effect of sludge dry solids to steam from bark boiler

Biosludge incineration in a bark boiler impacts the boiler operation. In this study, the impacts were studied using a calculation template. Table 8.10 presents the values used for bark, biosludge, and their mixture in the calculations. The mixture consists of 10% of biosludge and 90% of bark. The composition of bark does no change much after adding sludge, but one can notice that amounts of nitrogen, sodium, sulphur, chlorine and calcium increase slightly. Even if the increases are moderate, they can cause some problems in boilers. Especially alkali and chlorine corrosion risk has to be taken into account when biosludge is combusted.

	Unit	Bark	Biosludge	Mixture
С	m-%	53,9	50,4	53,5
Н	m-%	5,9	6,0	5,9
Ν	m-%	1,2	0,7	1,2
Na	m-%	1,5	9,7	2,3
S	m-%	0,7	3,7	1,0
Cl	m-%	0,4	2,5	0,6
Κ	m-%	0,15	0,16	0,15
Ca	mg/kg	0,3	920	92,2
HHV	MJ/kgds	20,88	20,77	20,87

Table 8.10. Compositions of bark, biosludge and their mixture.

Changes in steam production were estimated when the share and moisture content of biosludge varies. Moisture contents of 75%, 85% and 95% were used. The share of biosludge varied between 0% and 15%. The results are presented in Figure 8.26. For example, when sludge moisture content is 85% and its share in the fuel is 10%, the steam production decreases 2% compared with normal operation.

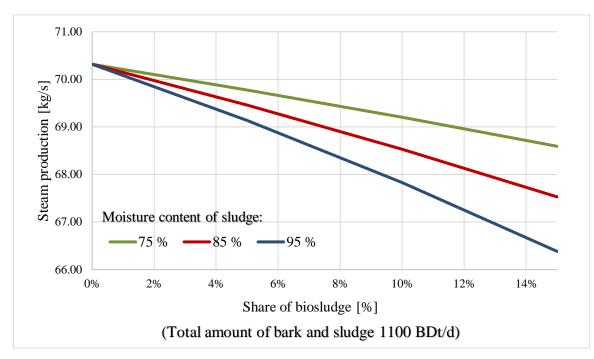


Figure 8.26. Example of the effect of adding biosludge to bark on steam production.

# 8.3 Effect of sludge dry solids to steam from recovery boiler

In this study, effects of biosludge incineration on the operation of a recovery boiler were studied using a calculation template. Table 8.11 presents the properties of biosludge, black liquor, and their mixture. Biosludge is dewatered for incineration to a dry solid content of 19.7%, which is relatively high for biosludge. There is 1% of biosludge and 99% of black liquor in the mixture. Biosludge addition does not change the properties of black liquor significantly due to the small share of sludge.

It was examined how the amount and moisture of the biosludge affect the operation of the recovery boiler by calculating the steam production of the boiler. Moisture contents of 75%, 85% and 95% were used. 85% is a typical moisture content of biosludge. In the calculations, it was assumed that the evaporator does not dry the mixture to normal dry solid content but the added biosludge decreases the dry solid content of the feed black liquor.

	Unit	Biosludge	Black liquor	Mixture
С	m-%	50.4	33	33.2
Н	m-%	6.0	3.2	3.2
Ν	m-%	0.7	0.1	0.1
S	m-%	3.7	5.2	5.2
Cl	m-%	2.5	0.3	0.3
Na	m-%	9.7	20	19.9
Κ	m-%	0.16	1.4	1.4
Ca	mg/kg	920	-	9.2
Р	mg/kg	0.2	-	-
Mg	mg/kg	230	-	2.3
HHV	MJ/kgds	20.77	13.6	13.7
S/(Na <sub>2</sub> +K <sub>2</sub> )	mol-%	54.2	35.8	36.0
$Cl/(Na_2+K_2)$	mol-%	6.6	0.9	1.0
K/(Na+K)	mol-%	1.0	4.0	4.0
Dry solids	%	19.7	80	79

Table 8.11. Properties of biosludge, black liquor and mixture. (Vakkilainen & Pekkanen, 2002)

Behavior of steam production with different biosludge moisture contents and shares is presented in Figure 8.27. The black liquor dry solids content was 85%. The decrease of steam production was found to be moderate. When the share of biosludge is 1%, the steam production changes for moisture contents of 75%, 85%, and 95% are 0.16%, 0.31%, and 0.47%, respectively. The effect on the recovery boiler's operation does seem small, but in a long run, the amount of lost steam is notable. For example, if the capacity of the recovery boiler is 3000 tds/d and 1% of black liquor is replaced with biosludge at moisture content of 85%, the loss of steam is about 27 000 kg/d.

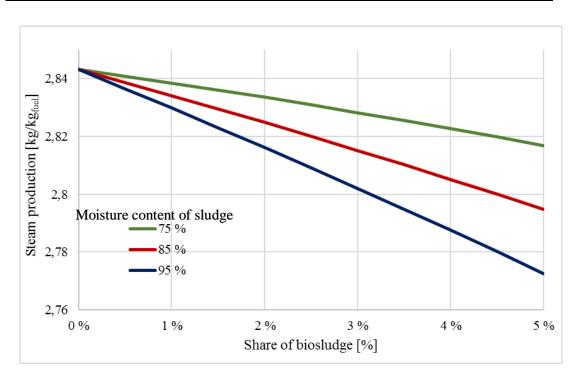


Figure 8.27. Effect of sludge incineration on steam production of an example recovery boiler.

# 9 Economics

The economics of the biosludge treatment system is affected by several factors. The disposal is closely linked to the other operations of the mill, and therefore, local conditions affect the feasibility of the processes, both technically and economically. Because sludge cannot be disposed at landfill anymore it has become imperative to find an economical alternative solution. The suggested new processes, such as anaerobic gas production or HTC treatment of sludge, affect the energy balances of the mill, as also does the commonly used conventional disposal method, incineration of relatively wet sludge in the mill's boilers. The economic feasibility of the disposal methods depends on the operational environment, such as local regulations related to waste management, and mill-specific details, such as financial significance of produced heat and electricity.

The costs of a sludge disposal process consist of investment cost including auxiliary equipment, such as pipes, possible storages, and drying equipment if the solid product fraction is further dried before combustion or other end-use. Operational costs include labour, maintenance, and energy use, the value of which is dependent on adequacy of it at the mill and on its possible sale price. Since landfill disposal is no longer possible due to regulation for example in Finland, the studied concepts should be compared with other possible treatment or end-use means. The benefits of new concepts include the avoided cost of other sludge treatment process.

A pulp mill site can be a favourable choice for an HTC or biogas installation due to existing infrastructure, such as wastewater and -gas treatment facilities and because of existence of suitable sources and sinks of heat. The costs of the studied concepts depend on the fitting to existing processes, layout and the needed additional equipment, which require mill-specific decision-making.

In addition to the above-mentioned utilization possibilities, Nuotio (2018) presented a techno-economic evaluation on utilization of biosludge as a fertilizer after thermal drying and granulation. According to the evaluation, the process seems feasible and could offer

a possibility to additional revenue if realized. Further research and piloting is however needed to get a realistic view on this possibility.

## 9.1 Economic evaluation of reference mill HTC cases

The feasibility of the HTC integration concepts presented in section 8 is roughly evaluated by estimating the investment cost and the possible profit. The investment cost estimates are based on the previous cost analysis of HTC of Nordic forest biomass by Saari et al. (2016). The cost of a reference facility was calculated based on the assumption that the size of the process components scales linearly with the mass flow ( $\dot{m}$ ) of the wet feed. The total capital investment (*TCI*) for each studied reference mill was calculated based on the cost of the reference facility as follows:

$$TCI = TCI_{ref} \left(\frac{\dot{m}_{\text{sludge}}}{\dot{m}_{\text{sludge,ref}}}\right)^{0.67}$$
(3)

The estimate includes the equipment for HTC and hydrochar after-treatment in each case. The preceding mechanical drying is not included, since in the reference cases mechanical drying existed also in the 0-case. It is, however, common that biosludge and primary sludge are mixed and dried simultaneously. When HTC is used only for biosludge, a separate drying equipment may be needed.

The investment costs for each mill case are collected in Table 9.1. The HTC process is still in the development stage and no large facilities exist; there are several uncertainties related to the economics of the concept. Therefore, the results should be considered as best estimates and used with care.

Table 9.1. Estimated investment cost (M€) for each HTC case in the reference mills.

	Base	OptA	OptB
Euca	15	15	17
NorPulp1	7	7	8
NorPulp2	9	9	10
NorInt1	10	11	12
NorInt2	15	15	17

To evaluate the economic feasibility of the studied cases, the payback period was calculated for each case using the investment estimates above and the profit that can be gained from additional electricity sales. 20-year lifetime, 6% interest rate, and operational and management cost of 3% of the investment were chosen for the calculations. Figure 9.1 shows the payback period as a function of electricity sale price for the studied cases when only income from sold electricity is taken into account. Using the assumptions above, for some of the cases the payback period exceeds 20 years within the studied electricity prices; these are not depicted in the figure. To make the cases NorPulp1 and NorInt1 and the HTC base case for NorInt2 feasible in the assumed conditions, additional profit from HTC is needed.

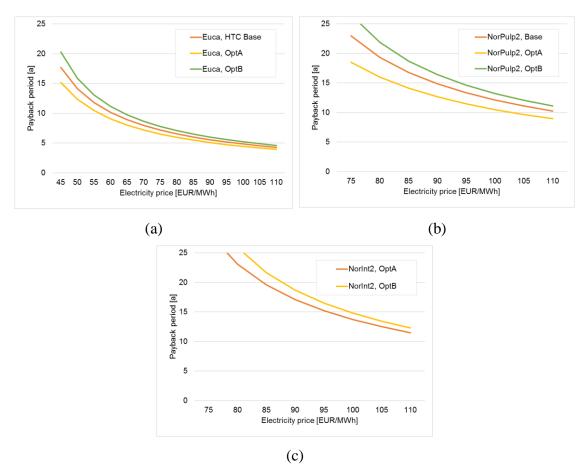


Figure 9.1. Payback period for the studied HTC integration cases in reference mills Euca (a), NorPulp2 (b), and NorInt2 (c).

The calculated payback periods are greatly dependent on electricity prices. Electricity prices have recently been in a turmoil due to the coronavirus pandemic causing falling demand and the structural changes in the power generation, namely, the decrease of fossil fuels and increase of especially solar generation. The second quarter of the year 2020 showed exceptionally low electricity prices; in Europe, the average price of the nine major markets was 19 €/MWh, which is 52% lower than in the second quarter of 2019 (European Commission, 2020). The divergence was however high, because of several different factors, such as level of lockdown measures and weather conditions, which affected the price development. Variation in the prices can be expected also in the future, due to for example the increasing presence of renewable power.

### 9.2 Economic evaluation of reference mill biogas cases

Anaerobic digestion of sludge offers several economic benefits for a pulp and paper facility, such as the possibility to dispose of sludge on site without further transportation costs, possible income from biogas sale or use of biogas as a substitute for purchased (fossil) fuel, and use for excess low temperature heat streams. Additional income is possible if the digestate can be sold for soil amendment. The costs of the digestion process consist mainly on the investment and maintenance. In an on-site process, maintenance may be merged with the other mill operations.

The feasibility of the anaerobic digestion in reference mill cases is estimated based on recent projects and a previous study (Nuotio, 2018). The total capital investment for each mill was calculated similarly as for the HTC cases above. Payback period was calculated for each mill case using the investment estimates above and the profit that can be gained from additional electricity sales. For this evaluation, it was estimated that biogas could be used to substitute fossil fuels in the lime kiln, namely oil or natural gas. The income of the process then consists of save in fossil fuel price and the sale of additional electricity. The price of fossil fuels is estimated based on recent price development. The yield and properties of biogas from anaerobic digestion depend on the properties of feed and process conditions. The possible energy recovery is here estimated at 1.7 MWh/tds based

on the results of Stoica et al. (2009). This can however be considered only an estimate since local conditions vary greatly. Table 9.2 presents the basis for the economic evaluation and Figure 9.2 shows the payback period for each case. 20-year lifetime, 6% interest rate, and operational and management cost of 3% of the investment were chosen for the calculations.

	Unit	Euca NorPulp		NorPulp2	NorInt1	NorInt2	
Investment	M€	12.5	4.0	6.3	7.0	11.1	
Lime kiln fuel	-	Oil	Oil	Oil	NG	NG	
Lime kiln fuel price	€/MWh	58	58	58	48	48	
Biogas share of lime kiln energy	%	4.7%	1.6%	3.3%	1.7%	3.3%	

Table 9.2. Basis for the economic evaluation.

Payback period is shown in Figure 9.2 as a function of electricity price. Lower than 10 years payback period is reached for mill cases Euca, NorPulp2, and NorInt1 when the price of electricity rises above 35, 75, and 90 €/MWh, respectively. Since biogas is used to replace fossil fuels, fossil fuel price level affects the payback period. A rough sensitivity analysis using the electricity price range in the figure shows that 50% increase in oil/NG prices would give a payback period at or below 10 years for all the other mill cases than NorPulp1. The results are greatly dependent on the electricity prices; recent electricity prices were discussed in the previous section 9.1.

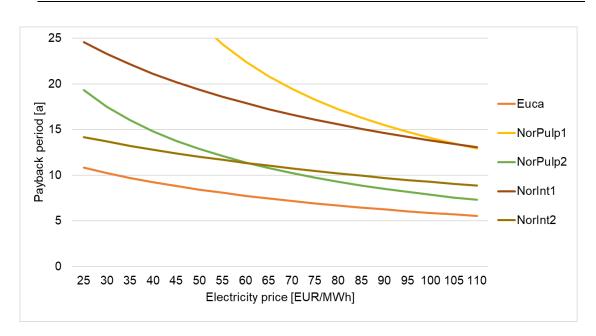


Figure 9.2. Payback period for biogas production from sludge in the reference mills.

### 9.3 Utilization possibilities of hydrochar

Section 8 presented the calculations on the effect of HTC process in the viewpoint of energy consumption and production. The calculations show that HTC integration in a pulp mill brings benefits in the form of increased energy generation when HTC-treated biosludge is incinerated in the mill boilers as an option for incineration of untreated biosludge. Also, when biosludge is incinerated in the recovery boiler it increases the boiler load. If the recovery boiler is the bottleneck of the pulping process, biosludge disposal using other methods enable pulp production increase,

HTC treatment offers, however, also other utilization possibilities for biosludge in addition to incineration. HTC has previously been presented as a method to produce for example activated carbon, and HTC-treated biomass can be used as soil conditioner, unlike untreated biosludge. Biosludge typically includes e.g. nitrogen and phosphorus that are needed in fertilizers. The amount of harmful compounds such as mercury (Hg), should be taken into account when further utilization possibilities of hydrochar are considered. Depending on the properties of biosludge, several possibilities can be found.

Recently, carbon balances of the industrial units have been under lively discussion due to increased concern on climate change and its implications. Pulp mills among other industrial operators are looking for options to reduce carbon dioxide emissions. In the case of a modern pulp mill, relatively small amount of carbon capture and storage could make a pulp mill a carbon sink instead of a carbon source (Kuparinen et al., 2019). If HTC-treated biomass is used as soil conditioner, the carbon in the biosludge is then stored and not released in the atmosphere. For a pulp mill, this can offer a way to get closer to negative emissions.

### 9.4 Utilization possibilities of biogas

The anaerobic digestion of biosludge produces biogas that consists mostly of methane and carbon dioxide. The typical use for the gas is combustion in a boiler or using a gas burner to produce heat and/or electricity. In a kraft pulp mill, a relatively easy utilization option is to use the gas in lime kiln, which typically is the primary fossil fuel user of the mill. Biogas use can partly substitute for fossil fuels in the kiln and is then among the means for more sustainable operation. In many mills, alternative fuels are already cocombusted in lime kilns with oil or natural gas, which are the typical primary fuels. The existing equipment determines what kind of modifications, if any, are needed in the combustion processes if biogas use in lime kiln is started. Gas can also be upgraded to be used for example as vehicle fuel; in Sweden, this is done for 60% of gas produced at wastewater treatment plants (Puig von Friesen et al., 2018).

The anaerobic digestion process produces also digestate, that can be even more difficult to utilise than biosludge, even though its mass and volume are lower than those of the original sludge. The digestate can be mixed with primary sludge or biomass, such as grass, to make it easier to dewater, and then e.g. composted or used as fuel after suitable treatment. It can also be suitable for soil amendment in areas not used for food production; the typically high level of cadmium is a hindrance for use in food crop areas (Hagelqvist, 2013).

### 10 Summary

The wastewater treatment systems in the forest industry produce notable amounts of primary, secondary, and tertiary sludges with minor amounts of other sludges. These sludges are challenging to dispose of due to their properties. Particularly challenging is secondary sludge (biosludge), both because of its properties and because the recovered amount per ton of pulp has been increasing lately. Simultaneously, the legislation on waste management tightens and the demand for more environmental-friendly and energy-efficient production processes increases. These factors act as driving forces to find new ways for sludge, particularly biosludge, disposal. In industry tertiary sludge disposal as separate item has not been considered. This study compared the currently used disposal methods and presented potential new options via literature review and case calculations.

Currently, forest industry sludges are usually incinerated in boilers on site. Due to high moisture content of sludge, incineration does not often bring benefits in the form of produced energy but only acts as a disposal method. This, however, increases the needed boiler capacity and has often resulted in lowered availability and increased corrosion. This study focused especially on biosludge handling due to it being the most problematic sludge stream at the moment. Primary and tertiary sludge treatment and disposal is currently done mainly in biomass boilers. Biosludge is incinerated also in recovery boilers in some mills.

In the EU area, directives on waste and on landfill of waste control waste management. The primary aim is to prevent the generation of waste. Landfilling of biodegradable waste is prohibited. Reuse is set as the primary method for waste disposal, followed by recycling of materials and energy recovery. Forest industry sludges include e.g. nutrients and if suitable treatment processes can be found, their re-use instead of disposal would promote circular economy. Primary sludge forms in the primary clarifiers of a modern wastewater handling system. It consists of compounds that settle easily, such as fibres and other solids generating in pulping and papermaking.

Biosludge forms in biological water treatment systems when microbial mass agglomerates on the bottom of the pond. In the forest industry, biosludge is a challenging waste fraction due to its high moisture content and problematic properties that complicate dewatering and limit reuse possibilities. Currently, biosludge is often incinerated in the boilers of the mills. The heating value of biosludge is low, in some cases negative due to moisture content. Therefore, incineration of biosludge can be energy consuming instead of generating energy. Biosludge also includes components, such as chlorine, sodium, and potassium, that can harm the combustion equipment.

Biosludge and primary sludge are often mixed and handled together in the chemical forest industry to make biosludge handling easier. The increasing amounts of biosludge however lead to higher shares of biosludge in mixed sludge, which makes mixed sludge drying more difficult.

Tertiary sludge, also called chemical sludge, forms in the mills that have tertiary wastewater treatment process. Tertiary sludge consists mainly of flocs that are formed by the used coagulants in the treatment. A tertiary treatment process can be for example chemical precipitation followed by filtration or clarification to remove the formed flocs. The sludge composition depends on the dosage and the chosen chemicals. The amount of tertiary sludge is typically smaller than primary or secondary sludge, and tertiary treatment is only used in a part of the mills. Tertiary sludge handling methods vary greatly due to varying sludge properties and the lack of established processes.

The main problem in sludge disposal is the high moisture content. The EU Best Available Techniques (BAT) reference documents (BREFs) present the techniques and processes that are considered the best in terms of emissions and consumption levels. Biosludge handling is typically a two-stage process, where the first stage is thickening and dewatering and the second stage disposal. Although biosludge is often mixed with primary sludge to improve the dewatering properties, separate handling would offer more reuse possibilities. The fibre-rich primary sludge has more reuse possibilities than biosludge or tertiary sludge. Separate handling of sludges enables different utilization possibilities for different types of sludges and promotes utilization instead of incineration or disposal.

This study presented biogas production and hydrothermal carbonization (HTC) as new alternatives for biosludge treatment. Anaerobic digestion of biomass is a known process for biogas production, but its use for the handling of forest industry sludges is new. The properties of raw material affect the process and gas formation, and the high concentration of lignocellulosic biomass and varying properties of forest industry sludge streams complicate the process. Anaerobic digestion, however, offers several advantages in sludge handling. It reduces the volume and dry mass of sludge and produces biogas, that can be utilized e.g. for energy purposes. The digestate from the process can be used for land applications or, after a suitable treatment, incinerated for energy generation. The process has been trialled in one mill in Finland.

HTC is a thermochemical process where biomass is treated in a suspension of water to produce solid, coal-like product called hydrochar, and liquid and gaseous byproducts. HTC is especially suitable for wet biomasses because it does not require preceding drying. It has been successfully tested for various biomass types. Hydrochar is stable, non-toxic product, that is easier to handle than the original biomass. It can be used for several purposes, such as replacing coal in energy production applications or use as soil conditioner. HTC is a promising alternative, because it transforms sludge into usable product consuming only a moderate amount of energy in the process. The process will be trialled in one mill in Finland.

The feasibility of anaerobic digestion and HTC and their effect on the mill energy balances was estimated by calculating reference mill cases. The case studies compared the new treatment processes to the conventional situations, where sludge is incinerated in the mill boilers. The case studies show that although biosludge makes up a small portion of the fuel flow into the boilers, the studied new handling methods can improve the boiler operation and increase steam and electricity consumption of the mill. The economic feasibility depends greatly on local conditions and mill details.

### **11 Conclusions**

This study discussed and evaluated the different types of sludges in the processes of forest industry, how and where they form, and how they are handled, used, or disposed of now and in the future.

Currently, the dominating method for sludge disposal is incineration in the own boilers of the forest industry units. Incineration is an easy solution for sludge disposal in mills where boilers already exist, but it also has several disadvantages, including the need for higher boiler capacity, harmful effects to the combustion equipment due to corrosion causing components in biosludge, and low or even negative additional energy generation due to high moisture content of sludge. The prevailing trend today is circular economy and the endeavour for low or zero-waste production. Therefore, new processes are needed for sludge handling to promote utilization over disposal. The pulp and paper mills are likely to transform into forest biorefineries in the coming years, and in the variety of product lines of a future biorefinery, value-added chemicals, materials, and fuels from produced from mill side-streams, such as sludges, play a notable role.

This study presented biogas production and hydrothermal carbonization (HTC) as new alternatives for biosludge treatment. Both processes enable biosludge handling in a way that produces usable product with a moderate amount of energy. Both processes also are promising, but novel alternatives for sludge handling. Biogas production using anaerobic digestion process has several advantages, since it reduces the volume and dry mass of sludge and produces utilizable biogas and digestate, that can be used for example in land applications. The HTC process transforms biosludge into hydrochar and consumes moderate amount of energy in the process. Hydrochar can be used for energy purposes or e.g. as soil conditioner.

The case studies presented in this study showed that it is possible to economically apply a hydrothermal carbonization or anaerobic digestion process to produce hydrochar or biogas in a pulp mill under certain conditions. However, the economic feasibility of the processes requires careful consideration in each separate mill case. Based on the reference mill calculations, the payback period for HTC or anaerobic digestion process integrated in a kraft pulp mill can be at the lowest around five years. This implies, that it might be feasible to invest in a process when constructing a new mill, but not likely to add the process in an existing mill, where already exists and old sludge handling process. It should also be noticed that each mill case is different; local conditions, sludge properties and amounts of different types of sludges vary greatly. Therefore, a common recommendation for the best method for sludge handling cannot be given. The possible uses for products from sludge handling process are an important aspect when sludge handling processes are discussed. For example, when there is a need for a renewable lime kiln fuel, biogas may be a desired product, as well as when there is need for biogas for vehicle use in the neighbourhood.

A new and attractive commercial process does not exist yet even though mill tests for biogas and HTC have been done. Until they have gained wider acceptance, the destruction of biosludge by co-combustion in either biomass boiler or recovery boiler must be utilized.

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Mill		Α	B	С	D	E	av.	min.	max.
Dry solids content	%	9	3	12	0.97	2.4	5.5	1.0	12.0
Dry solids composi									
Ash (575 °C)	%	16	24	13	38	29	24.0	13.0	38.0
Ash (815 <sup>0</sup> C)	%	15	24	-	37	25	25.3	15.0	37.0
С	% ds	44.2	38.6	46.2	31.9	37.2	39.6	31.9	46.2
Н	% ds	6	5.1	6.4	3.8	5	5.3	3.8	6.4
Ν	% ds	6.7	3.4	5.3	3.6	4.2	4.6	3.4	6.7
Na	g/kgds	7.3	20	5.8	60.1	37	26.0	5.8	60.1
K	g/kgds	2.8	2.9	1.7	5	1.5	2.8	1.5	5.0
S	g/kgds	12.2	25	17.6	37	16	21.6	12.2	37.0
Ca	g/kgds	17	26	17	21	41	24.4	17.0	41.0
Si	g/kgds	21	17	9.9	36	23	21.4	9.9	36.0
Cl	g/kgds	3.5	4.4	3.5	3.5	51	13.2	3.5	51.0
Al	g/kgds	3.3	23	3.7	17	6.3	10.7	3.3	23.0
Fe	g/kgds	3.2	6.9	3	23	3.5	7.9	3.0	23.0
Mg	g/kgds	11	6.3	2.1	7.1	10	7.3	2.1	11.0
Р	g/kgds	7.9	5.4	4.6	5.9	5.6	5.9	4.6	7.9
Mn	mg/kgds	6400	8300	1070	2100	7000	4974	1070	8300
Zn	mg/kgds	300	920	630	95	710	531.0	95.0	920.0
Ba	mg/kgds	550	460	-	200	300	378	200	550
Ti	mg/kgds	120	180	-	440	170	228	120	440
V	mg/kgds	< 10	67	-	35	34	45.3	34.0	67.0
Cr	mg/kgds	60	45	29	29	57	44.0	29.0	60.0
Ni	mg/kgds	53	48	13	19	70	40.6	13.0	70.0
Cu	mg/kgds	36	38	37	42	26	35.8	26.0	42.0
F	mg/kgds	52	32	0	58	< 25	35.5	0.0	58.0
Pb	mg/kgds	7.9	40	13	7.4	15	16.7	7.4	40.0
Со	mg/kgds	< 4	16	21	< 4	10	15.7	10.0	21.0
Cd	mg/kgds	8	5	6	0.7	8.6	5.7	0.7	8.6
As	mg/kgds	5.6	3	-	4.6	2.2	3.9	2.2	5.6
Sb	mg/kgds	< 0,5	0.5	-	< 0,5	< 0,5	0.5	0.5	0.5
Hg	mg/kgds	< 0,1	< 0,1	0.6	< 0,1	0.1	0.4	< 0,1	0.6
T1	mg/kgds	0.45	0.08	-	0.26	0.32	0.3	0.1	0.5
Se	mg/kgds	< 0.2	< 0.2	-	< 0.2	< 0.2	< 0.2	0.0	< 0.2
HHV, dry solids	MJ/kgds	19.03	16.1	-	13.88	15.6	16.2	13.9	19.0
LHV, dry solids	MJ/kgds	17.34	14.2	-	11.88	14	14.4	11.9	17.3
LHV, sample	MJ/kg	-0.58	-1.94	-	-2.3	-2.05	-1.7	-2.3	-0.6

# Appendix A: Biosludge properties

Mill		Α	В	C	D	E	F*	max.	min.	av.
Туре		mix	bio	bio	bio	bio	bio			
Mechanical dewater				Yes	No	Yes				
Dry solids (wt-%)	%	10.7	16.2	11	3.9	9.4	0.6	16.2	0.6	8.6
HHV (DS)	MJ/kg	11.1	15.2	13.7	11.6	16.3	11.7	16.4	11.2	13.3
LHV (DS)	MJ/kg	9.23	13.3	11.9	9.76	14.5	9.41	14.6	9.23	11.4
LHV (AR)	MJ/kg	-1.19	0.12	-0.86	-1.97	-0.84	-2.37	0.12	-2.37	-1.2
Composition	as DS									
Ash	%	17.1	10.2	16.2	16	17	51.9	51.9	10.2	21.4
Na	g/kg	22.7	10.3	9	15.3	4	89.2	89.2	4	25.1
С	%	45.6	52	44.8	46.7	45.8	29.4	52	29.4	44.1
Н	%	5.9	6.3	5.7	5.3	6	3.6	6.3	3.6	5.5
N	%	4.5	5.6	6.1	4.5	5.8	2.7	6.1	2.7	4.9
S	g/kg	20.1	18.2	16.9	23	14.7	48.3	48.3	14.7	23.5
К	g/kg	2.8	0.7	0.4	0.04	N.F.	3.4	3.4	0.04	1.5
Cl	g/kg	2.4	7.4	5.6	7.9	4.5	39.6	39.6	2.4	11.2
0	%	35.4	30.3	33.5	33.2	33.7	24.4	35.4	24.4	31.8
Al	g/kg	7.1	3.7	4.5	4.8	7	3.5	7.1	3.5	5.1
Zn	g/kg	0.6	0.09	0.3	0.6	0.8	0.3	0.8	0.09	0.4
Pb	mg/kg	18	11	14	14	16	12	18	11	14.2
Ca	g/kg	9.1	12.7	25.3	26.5	13.4	34.1	34.1	9.1	20.2
Mg	g/kg	4.2	2	8	6.3	4.6	3	8	2	4.7
Mn	g/kg	4.7	6.6	6	6.2	8.9	2	8.9	2	5.7
Р	g/kg	5.8	6	6.5	3.9	6.6	5.1	6.6	3.9	5.7
Fe	g/kg	15.1	1	3.4	4	6.9	28.9	28.9	1	9.9
Sn	mg/kg	< 15	< 15	< 15	< 15	< 15	< 15	< 15	< 15	< 15
Si	g/kg	6.6	2.8	17	10.5	18.1	7.6	18.1	2.8	10.4
SO <sub>4</sub> =	g/kg	0.9	1.3	0.8	0.8	1.3	143	143	0.8	24.7
CO <sub>3</sub> =	g/kg	4.3	3.4	12.1	16	10.5	39.8	39.8	3.4	14.4

## **Appendix B: Sludge analysis results**

\*Sludge F contains RB ESPash

#### SUOMEN SOODAKATTILAYHDISTYS RY, RAPORTTISARJA

- 1/2019 Suomen Soodakattilayhdistys ry Konemestaripäivä 24.1.2019, esitelmät Scandic Laajavuori, Jyväskylä, Metsä Fibre Oy, Äänekosken Biotuotetehdas (16A0913-E0194) 24.1.2019
- 2/2019 Suomen Soodakattilayhdistys ry Soodakattila-alan yhteistoiminta Vuosikertomus 2018 (16A0913-E0195) 11.4.2019
- 3/2019 Suomen Soodakattilayhdistys ry Soodakattila-alan yhteistoiminta Pöytäkirja. Vuosikokous 11.4.2019, Lapland Hotels Bulevardi, Helsinki (16A0913-E0196) 11.4.2019
- 4/2019 Suomen Soodakattilayhdistys ry Soodakattilapäivä 31.10.2019
   Sokos hotelli Presidentti, Helsinki (16A0913-E0197) 31.10.2019
- 5/2019 Suomen Soodakattilayhdistys ry Mustalipeän polttomenetelmät Suomen soodakattiloissa 2018 Esa Vakkilainen, LUT-yliopisto (16A0913-E0198) 1.11.2019
- 6/2019 Suomen Soodakattilayhdistys ry Non-process elements in the recovery cycle of six Finnish kraft pulp mills Camilla Karlemo, Åbo Akademi (16A0913-E0199) 1.11.2019
- 7/2019 Suomen Soodakattilayhdistys ry Evaluation of three different gas phase chemistry mechanisms for predicting NOx emission formation in recovery boilers Markus Engblom, Åbo Akademi (16A0913-E0200) 1.11.2019

#### 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