



**ENVIRONMENTAL PERFORMANCE OF PULP AND PAPER MILL BIOSLUDGE
UTILIZATION BY HYDROTHERMAL AND BIOCHEMICAL TREATMENT**

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ABSTRACT

Sludge is the largest solid waste stream generated in pulp and paper mills. Biosludge, which accounts for 30% of the total amount of sludge generated, is difficult to handle mainly due to a moisture content over 90%. While the most common end-of-life treatment for biosludge in Finland is combustion, this option is not efficient in recovering the energy and nutrients contained in biosludge. This calls for alternative treatment methods. Therefore, this study carried out a comparative environmental life cycle assessment (LCA) of four different treatments: hydrothermal carbonization (HTC) to produce hydrochar for energy or nutrient recovery, anaerobic digestion (AD) combined with HTC, separate hydrolysis and fermentation (SHF). Combustion of biosludge was considered as the baseline scenario. The LCA results show that HTC and AD technologies improve the environmental performance of biosludge handling in four out of seven impact categories. HTC performance could be improved by optimising the valorisation of the filtrate. SHF results in considerably higher environmental impacts than combustion and other alternative technologies in several impact categories, mainly because of enzyme production. However, SHF of biosludge is still under development, leading to high uncertainties on the data.

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SYMBOLS AND ABBREVIATIONS

Abbreviations

AC	Ash content
AD	Anaerobic digestion
BCEF	Biogenic carbon equivalency factor
BMP	Biochemical methane potential
BOD	Biochemical oxygen demand
BS	Biosludge
CC	Climate change (including biogenic carbon)
CE	Cellulase enzyme
COD	Chemical oxygen demand
DS	Dry solids
FC	Fixed carbon
FE	Freshwater eutrophication
FPMF	Fine particulate matter formation
FU	Functional unit
GHG	Greenhouse gas
HC	Hydrochar
HHV	Higher heating value
HTC	Hydrothermal carbonization
HTc	Human toxicity (cancer)
HTR	Hydraulic retention time
LCA	Life cycle assessment

LHV	Lower heating value
MC	Moisture content
ME	Marine eutrophication
P&P	Pulp and paper
SRT	Solids retention time
TA	Terrestrial acidification
TET	Terrestrial eco-toxicity
VM	Volatile matter
WWTP	Wastewater treatment plant

Table of contents

Abstract

Acknowledgements

Symbols and abbreviations

1. Introduction9

LITTERATURE REVIEW

2. Biosludge production and challenges in pulp and paper industry 11

2.1. Paper mill wastewater treatment 11

2.2. Properties of biosludge..... 13

2.2.1. Proximate composition 15

2.2.2. Elemental composition 16

2.2.3. Inorganic elements 17

2.2.4. Heating value 18

2.3. Challenges of biosludge handling 19

3. Biosludge conversion technologies20

3.1. Thermochemical conversion of biomass.....21

3.2. Direct incineration of biosludge.....23

3.3. Hydrothermal carbonization (HTC).....27

3.3.1. Process27

3.3.2. Hydrochar properties and applications28

3.3.3. Filtrate properties and applications.....30

3.4. Anaerobic digestion (AD).....31

3.4.1. Process31

3.4.2. Biogas production35

3.4.3. Digestate properties and applications37

3.5. Fermentation37

3.5.1. Process38

3.5.2. Application.....40

METHODOLOGY

4. Life cycle assessment of pulp and paper mill biosludge treatment	41
4.1. Goal and scope definition.....	41
4.2. Inventory analysis	44
4.2.1. General assumptions	44
4.2.2. System 0 - Incineration	45
4.2.3. System 1 - HTC + incineration	47
4.2.4. System 2 - HTC + soil amendment.....	48
4.2.5. System 3 - AD + HTC + Soil amendment	48
4.2.6. System 4 - Fermentation	49
4.2.7. System expansion	50
4.3. Life cycle impact assessment	52
4.4. Uncertainty analysis	53
RESULTS AND DISCUSSION	
5. Results	55
5.1. Contribution analysis	57
5.2. Sensitivity analysis.....	60
6. Discussion	62
7. Conclusion	68
References.....	69
APPENDIX.....	81

Appendices

Appendix 1. Life Cycle Inventory for system 0.

Appendix 2. Life Cycle Inventory for system 1.

Appendix 3. Life Cycle Inventory for system 2.

Appendix 4. Life Cycle Inventory for system 3.

Appendix 5. Life Cycle Inventory for system 4.

Appendix 6. Life Cycle Inventory for the boiler and flue gas control of every scenario.

Appendix 7. LCIA results and contribution analysis.

Figures

Figure 1. Pollutants released from various steps of paper manufacturing process.....	12
Figure 2. Wastewater treatment plant in pulp and paper industry.....	13
Figure 3. Schematic model of various forms of water in biosludge.....	15
Figure 4. Biomass conversion technologies.....	21
Figure 5. Development of the elemental composition of biomass during HTC.....	29
Figure 6. Schematic representation of anaerobic digestion process.....	32
Figure 7. Process overview of lignocellulosic bioethanol production via enzymatic hydrolysis and fermentation.....	39
Figure 8. Life cycle diagrams.....	42
Figure 9. LCIA contribution assessment of each scenario compared to the baseline scenario (S0).....	57
Figure 10. LCIA contribution assessment for the selected impact categories.....	58
Figure 11. Influence of biogenic carbon equivalence factor on climate change.....	63
Figure 12. Climate change contribution assessment excluding bark and fines treatment, biogenic carbon emissions and modelling system expansion with co-products displacement.....	65

Tables

Table 1. Biosludge characterisation.....	14
Table 2. Maximum heavy metal content for sludge utilization as a fertilizer in Finland	18
Table 3. Comparison of reaction conditions and typical product yields for thermochemical conversion processes.....	22
Table 4. Lignocellulosic content of different feedstocks.....	33
Table 5. Characteristics of feedstocks for combustion.....	45
Table 6. Inventory related to system expansion.....	51
Table 7. Selected impact categories of the ReCiPe 2016 methodology.....	53
Table 8. Sources of uncertainties in this study.....	54
Table 9. Key parameters chosen for the sensitivity analysis.....	55
Table 10. LCIA results.....	56
Table 11. Sensitivity ratio for the selected parameters.....	61

1. Introduction

Pulp and paper (P&P) industry is currently in a transition phase to switch from a linear model, where most of the waste is incinerated, to a circular approach where mills are becoming biorefineries upgrading each waste streams into resources with high energy, nutrient, and carbon sequestration potentials (Kinnunen et al., 2015). This transition is operating all around the world and particularly in Finland where the industry is of great importance in the economy of the country. Finland is the second largest pulp producer of Europe, after Sweden (CEPI, 2022), it exports 1.5 billion euros of pulp annually and paper and paper board account for 5.7 billion euros of exports (Ministry of Agriculture and Forestry of Finland, 2022). Worldwide, 192 million tonnes of pulp for paper were produced in 2021 (Food and Agriculture Organization of the United Nations, 2023). Considering that each ton of pulp requires 35 tonnes of water to be produced (Ma et al., 2018) and generates around 50 kg of dry sludge (Oda et al., 2021), the environmental impacts of P&P industry are considerable. In the objective of integrating circular economy model in P&P mills, one of the waste streams that has drawn the attention of the scientific community for the past few years is secondary sludge (referred as biosludge) from pulp and paper wastewater treatment. While the most common disposal methods worldwide for biosludge (BS) are landfill and incineration, the physical and chemical properties of this waste stream make those treatments economically unsuitable and environmentally harmful. The high organic carbon content of biosludge (around 50 wt% dry basis) may cause leaching and greenhouse gas emissions in landfill (Hämäläinen et al., 2022), and the high moisture content and high volatile content result in a low heating value during incineration (Martinez et al., 2021). In this context, hydrothermal and biochemical treatments, such as hydrothermal carbonization, anaerobic digestion and fermentation, could help the P&P industry reducing its environmental impacts, energy consumption and increasing its cost-effectiveness.

Hydrothermal carbonization (HTC) is a relatively recent thermochemical process that consists in heating biomass in a aqueous environment to produce value-added resources. From this process, two main resources are produced: hydrochar (HC), a solid material with a high calorific value and rich in carbon, and a filtrate with high nutrient content. Hydrochar can be used for energy or nutrient recovery (Mohammadi et al., 2019a). As for the filtrate, it requires further treatment before being discarded. It can be directed to wastewater treatment

plant (WWTP) or recirculated to the HTC reactor for feedstock dilution, thus improving hydrochar properties (Banks, 2021). While HTC has attracted considerable attention recently, most of the studies focus on municipal sewage sludge as a feedstock and hydrochar utilization as an application (Cebi et al., 2022; L. Wang et al., 2019). Only few studies have analysed the environmental performance of HTC treatment of biosludge from P&P mills (Hämäläinen et al., 2022; Mannarino et al., 2022; Mohammadi et al., 2019a). Besides, to the author's knowledge, those existing studies have not yet compared the environmental impacts of HTC treatment with biochemical processes like anaerobic digestion (AD) and fermentation of biosludge. Anaerobic digestion is a microbial-mediated biological process in which biomass is converted into two valuable products: biogas and a fertilizer, called digestate. Despite its double potential, the treatment method is facing different technical challenges, mainly related to the instability of the process and presence of inhibitors in the feedstock that reduce yield and quality of the products (Pecchi & Baratieri, 2019). On the other end, fermentation is a biological process that converts sugars contained in biomass into bioethanol. Those sugars need to be released beforehand through hydrolysis to make the fermentation possible and produce the desired biofuel (Praveen Kumar et al., 2020).

Life cycle assessment (LCA) is a standardized method that evaluates the environmental impacts of a product, process or service through a detailed analysis over their respective life cycles. It has been used in many studies to assess the performance of different treatment methods of biosludge in P&P industry: landfill, incineration, HTC, AD only, AD followed by thermal treatment of the digestate (Mohammadi et al., 2019a, 2019b). But none of those has analysed the combination of AD and HTC of the digestate and compared it with classic HTC or fermentation processes.

To close the gap in the research, this study aims at assessing the environmental performance of combined hydrothermal and biochemical treatment of P&P biosludge, compared to the commonly practiced incineration. Five systems were analysed, using LCA methodology: (S0) Incineration of biosludge with energy recovery; (S1) HTC of biosludge followed by incineration of hydrochar; (S2) HTC of biosludge followed by hydrochar use as soil amendment; (S3) Anaerobic digestion of biosludge followed by HTC of digestate and hydrochar use as soil amendment; (S4) Hydrolysis and fermentation of biosludge. The objective is to compare the impacts and potentials of each treatment method and identify further improvements for the P&P industry. The type of sludge analysed is secondary sludge

(biosludge) from pulp and paper mills in Finland. After presenting the background information about biosludge characteristics and potential treatment methods, this study follows the four-step process of LCA methodology: goal and scope definition, inventory analysis, impact assessment and results interpretation (SFS-EN ISO 14040: 2006).

2. Biosludge production and challenges in pulp and paper industry

The papermaking process is highly energy and water intensive, which raises many environmental concerns (Dutt et al., 2021). To produce one ton of paper pulp, 35 tonnes of water are required (Ma et al., 2018). During the production process, this amount of water is getting polluted by the various raw materials and chemicals used (wood, gums, glues, colorants...) (Martinez et al., 2021). Considerable amount of sludge is then generated, 40 kg to 50 kg of sludge per one ton of paper produced, from which approximately 30% is biosludge (Oda et al., 2021).

2.1. Paper mill wastewater treatment

The four main processes of a typical integrated pulp and paper mill are raw material preparation, pulping, bleaching, and paper making. In each process, wastewater is generated, containing high concentration of pollutants. Pulp bleaching is responsible for most of the wastewater colour, organic matter, and toxicity (Dutt et al., 2021). **Figure 1** presents the effluent pollutants generated in each step of papermaking process, as well as wastewater volume and quality.

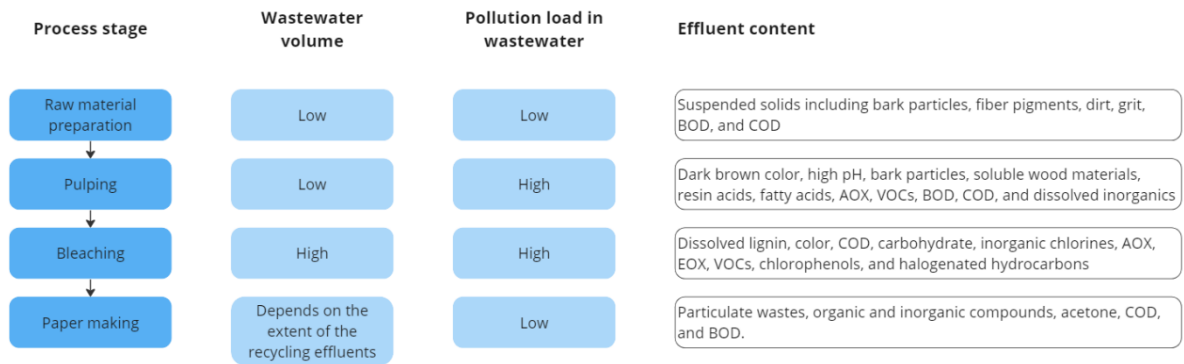


Figure 1. Pollutants released from various steps of paper manufacturing process. BOD: Biochemical oxygen demand; COD: Chemical oxygen demand; AOX: Absorbable organic halides; VOC: Volatile organic compound; EOX: Extractable organic halogens (Adapted from: Dutt et al., 2021; Kamali & Khodaparast, 2015; Pokhrel & Viraraghavan, 2004).

Wastewater from pulp and paper industry contains high organic substances, resulting in high biochemical oxygen demand (BOD) and chemical oxygen demand (COD). It also contains resin acids, chlorinated organics (absorbable organic halides), suspended solids, metals, fatty acids, lignin etc. (Dutt et al., 2021). Due to high levels of pollutants in effluent, pulp and paper wastewater can be harmful to aquatic ecosystems. Wastewater treatment is therefore essential to remove as much pollutants as possible before releasing effluents to waterways.

A wastewater treatment plant (WWTP) is mainly composed of three treatment stages, in which mechanical, biological and chemicals methods are used, as shown in **Figure 2**. After pre-treatment, wastewater is first going to a primary clarifier where effluent is treated chemically. In this stage, sedimentation is commonly used to remove suspended solids. Solids settle down at the bottom of the tank, forming primary sludge, which is then removed. Primary sludge is mainly composed of wood fibres (cellulose and hemicellulose fibres), papermaking fillers, pitch, by-products of lignin. Overflow is going to secondary treatment, where biodegradable organic materials are removed. This second step commonly uses activated sludge process which features an aeration tank and secondary clarifier, where wastewater is treated biologically, using micro-organisms. The later are using organic matter contained in wastewater and oxygen provided in the aeration tank as nutrition and generate carbon dioxide and water. Secondary sludge is produced in this stage, it is composed of microorganisms, undigested organics, inorganic material, unsettled fibres from primary treatment, concentrated metals. Tertiary treatment phase, also called advanced wastewater treatment, is optional and depends on the effluent's quality. It may include disinfection, membrane processes and activated carbon adsorption. In pulp and paper mills, wastewater

is particularly coloured, this last stage is thus focusing on removing the colour of the effluent. Wastewater treatment thus generates two types of solid waste: primary sludge and secondary sludge (also called biosludge). Sludges are the most important solid waste streams generated at pulp and paper mills. The average primary sludge to secondary sludge ratio in Canadian pulp and paper mills is 70:30, however this ratio can vary consequently from one mill to another (T. Meyer & Edwards, 2014). In many wastewater treatment plants, primary and secondary sludges are dewatered and mix together for further treatment. (Banks, 2021; Dutt et al., 2021; Hämäläinen et al., 2022; Mäkelä et al., 2018; Martinez et al., 2021.)

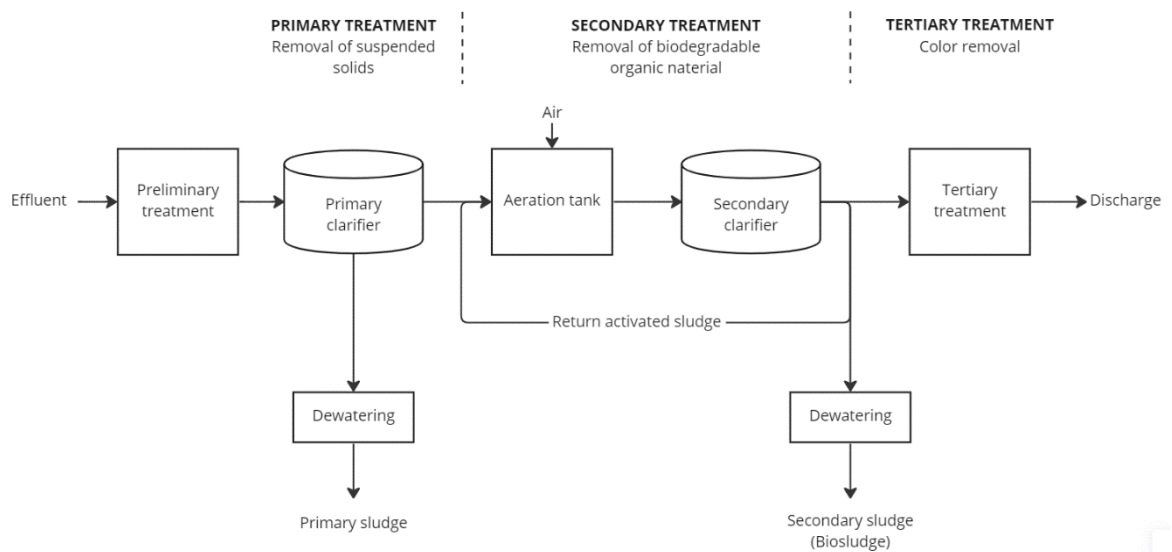


Figure 2. Wastewater treatment plant in pulp and paper industry (Dutt et al., 2021; Mäkelä et al., 2018; Martinez et al., 2021)

2.2. Properties of biosludge

Properties of biosludge varies from one mill to another, depending on raw materials, pulp and paper production processes, wastewater treatment procedures (Kinnunen et al., 2015). Typical biosludge characteristics from pulp and paper mills are presented in Table 1.

Table 1. Biosludge characterisation. DS: Dry solids.

Parameter	Biosludge composition	Unit
Proximate composition		
Moisture content (MC)	92.18 ^[1]	wt%
Ash content (AC) 550°C	20.99 ^[1]	wt% dry
Ash content (AC) 815°C	18.77 ^[1]	wt% dry
Volatile matter (VM)	69.16 ^[1]	wt% dry
Fixed carbon (FC)	12.06 ^[1]	wt% dry
Elemental composition		
C	44.76 ^[1] - 48.4 ^[2]	wt% dry
H	5.70 ^[2] - 5.80 ^[1]	wt% dry
O	23.6 ^[2] - 24.02 ^[1]	wt% dry
N	4.36 ^[1] - 7.9 ^[2]	wt% dry
S	2.30 ^[1] - 2.50 ^[2]	wt% dry
O/C ratio	0.49 ^[2] - 0.54 ^[1]	-
H/C ratio	0.12 ^[2] - 0.13 ^[1]	-
Heavy metals		
Cd	2 ^[3]	mg/kg DS
Cr	39 ^[3]	mg/kg DS
Cu	40 ^[3]	mg/kg DS
Ni	19 ^[3]	mg/kg DS
Pb	10 ^[3]	mg/kg DS
Zn	250 ^[3]	mg/kg DS
Other inorganic elements		
Na	0.42 ^[4]	wt% dry
Cl	0.00 ^[4]	wt% dry
K	0.15 ^[4]	wt% dry
Ca	1.13 ^[4]	wt% dry
Heating values		
Higher heating value (HHV)	19.78 ^[1] – 20.90 ^[2]	MJ/kg DS
Lower heating value (LHV)	18.5 ^[1]	MJ/kg DS
Lower heating value as received (LHVar)	0.6 ^[2]	MJ/kg DS

[1] (Martinez et al., 2021), [2] (Raatikainen, 2020), [3] (Mohammadi et al., 2019b), [4] (Oda et al., 2021)

2.2.1. Proximate composition

Proximate composition analysis is used to characterize moisture content (MC), volatile matter (VM), ash content (AC) and fixed carbon (FC) of a material. Samples are first dried for several hours at 105 ± 5 °C until constant mass according to EN 14774-2 (SFS, 2010). Proximate analysis then usually follows standard procedures DIN EN 15148:2010-03 (2010), DIN EN 14775:2010-04 (2010). Ash content is typically evaluated under standard temperature 550°C. Additional ash content analysis can be necessary in material containing high content of inorganic solids, like biosludge (Saari et al., 2022). In this case, temperature is increased to around 815 °C to analyse further ash content according to ISO 1171:2010.

Biosludge is mainly characterised by a high moisture content. Before dewatering process, biosludge moisture is over 90 wt% (Martinez et al., 2021). While dewatering is often required for further thermal treatment and volume reduction, this pre-treatment is particularly challenging for biosludge because of its gel-like structure and presence of extra-cellular polymeric substances, which bind water molecules to solid particles. According to (Mowla et al., 2013), there are four different forms of water in sludge, as shown on **Figure 3**. Free water is not attached to solid particles and can easily be removed by gravitational settling. Interstitial water is trapped within a floc structure or a cell, its removal by mechanical dewatering methods has low efficiency. Vicinal water and water of hydration are bond to solid particles, respectively physically bond to the surface of particles and chemically bond to the particles. They cannot be removed mechanically. Only thermochemical dewatering is efficient for water of hydration.

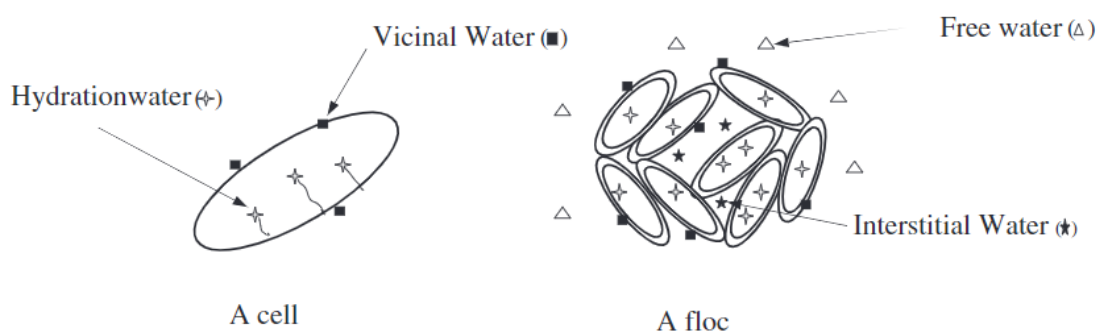


Figure 3. Schematic model of various forms of water in biosludge (Mowla et al., 2013)

Free water is the easiest form of water to be removed. The three other types, forming what is called bond water, require more energy to be dewatered. As around 80% of moisture content in biosludge is bond water, which cannot be removed using conventional mechanical dewatering methods, dewatering biosludge is both energy intensive and poorly efficient (Mowla et al., 2013). Usually, biosludge is mixed with primary sludge, thickened, and dewatered to reach a final dry solids (DS) content of 30-40 wt%. If biosludge is dewatered alone, DS content is less than 20%. (T. Meyer et al., 2018) analysed the benefits of mixing primary sludge with biosludge to improve biosludge dewatering. Only a small amount of primary sludge is necessary, up to 40% on a dry solid mass basis. This mix helps water circulation in sludge and thus facilitates dewatering.

The ash content of biosludge is also a characteristic of high concern, considering that it can contain harmful inorganic elements and is becoming residue after thermal treatment. The ash content of biosludge is approximately 17-21% DB, which is typically twice higher than in primary sludge (Martinez et al., 2021). Ash content in pulp and paper mill biosludge is influenced by the choice of raw materials. For instance, mills using virgin wood as raw material have lower ash content in their sludge than mills using recycled paper (Rissanen, 2020). Additional ash content analysis can be necessary in material containing high content of inorganic solids. In this case, the temperature is increased to around 815 °C to analyse further ash content (Saari et al., 2022).

2.2.2. Elemental composition

Elemental composition (C, N, H, S, O) of pulp and paper biosludge is given in **Table 1**. Carbon and oxygen are the main elements in biosludge because of the high fibre and organic matter. Hydrogen to carbon atomic ratio (H/C-ratio) and oxygen to carbon atomic ratio (O/C-ratio) are used as indicators of thermal energy potential for biomass. The lower they are, the higher the heating value (Martinez et al., 2021). Some thermochemical biomass conversion processes, like torrefaction, aim at increasing energy density of biomass by increasing its carbon content while decreasing hydrogen and oxygen contents (Basu, 2018). During combustion, nitrogen (N) and sulphur (S) contents can be problematic as they are converted into NO_x and SO_x, environmentally harmful and corrosive elements. The high concentration of nitrogen in biosludge, 14.2 times higher than in mineral soil at Northern Finland, might

potentially cause nitrate leaching if biosludge is directly used as a fertilizer or soil amendment (Pöykiö et al., 2018). The high content of sulphur in biosludge, between 2.3% and 2.5% of dry solids, is due to the used of Na_2S in kraft pulping process. The typical amount of sulphur in waste from wood (such as bark and fines) is below 0.02% on a dry basis (Saari et al., 2022).

2.2.3. Inorganic elements

Inorganic elements of biosludge, including heavy metals, are presented in **Table 1**. Those elements, present in ash, are raising several problems in biomass treatment and utilisation. Inorganic components, such as alkali chlorides, sulphates, carbonates and silicates, have to remain low for thermal treatment as they contribute to fouling and slagging during combustion of biomass (Praveen Kumar et al., 2020). Heavy metals are also limiting biosludge use as a fertilizer (Martinez et al., 2021). In Finland, regulations about sludge utilization as soil amendment are stricter than in most parts of the world. Maximum heavy metals contents are given in the Decree of the Ministry of Agriculture and Forestry on Fertiliser Products (Decree 24/2011). As shown on **Table 2**, all limits are respected for biosludge, except for cadmium (Cd) content. For all heavy metals analysed, their concentration in biosludge is still higher than in typical mineral soil in Finland, up to twenty times higher for zinc. Even if cadmium limit is not respected for use as a fertilizer, biosludge could be directly use as a soil improver, growing media or as a fertilizer in specific areas with different limitation, such as landfill sites in some industrial areas (Pöykiö et al., 2018).

Table 2. Maximum heavy metal content for sludge utilization as a fertilizer in Finland. All concentrations are expressed as mg/kg DS. [1] (Mohammadi et al., 2019b). [2] (Pöykiö et al., 2018). [3] (MMM, decree 24/11 2011).

Heavy metals	Typical amount in P&P biosludge [1][2]	Typical amount in mineral soil at Northern Finland [2]	Maximum content allowed [3]
Arsenic (As)	4.6	1.25	25
Mercury (Hg)	<0.04	0.01	1.0
Cadmium (Cd)	2 – 4.8	0.04	1.5
Chromium (Cr)	39 – 49	28	300
Copper (Cu)	20 - 40	17.5	600
Lead (Pb)	6.0 - 10	4.0	100
Nickel (Ni)	19 – 32	12	100
Zinc (Zn)	250 - 490	24	1500

2.2.4. Heating value

Heating value quantifies the energy released during combustion process. Lower heating value (LHV), or net heating value (NHV), is defined as the amount of heat released by the complete combustion of a unit of mass (initially at 25°C) once it has returned to its initial temperature less the heat of vaporization of water contained in the fuel (Basu, 2018). It considers the fact that temperature of flue gas is above the initial temperature of fuel, thus as water vapor in flue gas does not condense, latent heat of vaporization is not recovered. LHV is usually determined according to DIN EN 14918:2014-08 (2014) standard procedure and then calculated based on hydrogen content in biomass (Martinez et al., 2021).

Moisture and ash content are strongly correlated with biomass heating value. When moisture content is high, combustion process requires more energy to evaporate the water contained in the fuel. A high ash content means high inorganic elements or minerals content in biomass, which negatively impacts the calorific value. Higher heating value (HHV) is also correlated with high carbon and hydrogen contents. High carbon content typically implies high fixed carbon content which increases radiation heat transfer, however high fixed carbon content implies longer residence time to achieve complete combustion (Martinez et al., 2021). (Ozyuguran et al., 2018) studied 39 different biomass materials to predict the calorific value of biomass based on its ultimate composition. It concluded that linear equation containing two parameters, including carbon content and either nitrogen or oxygen content, makes it

possible to predict the calorific value of the material with a coefficient of determination (r^2) higher than 0.9219.

2.3. Challenges of biosludge handling

Sludge handling raises environmental, economic, and regulatory concerns. While the most common disposal methods worldwide are landfill and incineration, the physical and chemical properties of biosludge make those treatments unsuitable and environmentally inefficient. The high organic carbon content of sludge (around 50 wt% dry) may cause leaching and greenhouse gas emissions in landfill (Hämäläinen et al., 2022). For this reason, the EU has forbidden sludge disposal to landfill and gives the priority to energy recovery through incineration in the European Union Waste Framework Directive (2008/98/EC). However, the high moisture content and high volatile content result in a very low heating value. Thus, when biosludge is burned with other wood waste in mills, energy recovery potential is reduced. Some hazardous inorganics elements can also generate air, soil, and water pollution due to flue gas emissions and landfilling of ashes. Their presence limits as well biosludge use as a fertiliser (Martinez et al., 2021).

The environmental impacts and low efficiency of current biosludge disposal practices are motivating the P&P industry to find alternative methods. Biosludge conversion has different environmental advantages (Ding et al., 2021). It can reduce greenhouse gas emissions by decreasing mills' dependence on fossil fuels and increasing renewable energy share. It reduces resources depletion by creating new value-added products such as fertilizer and fuel. Avoiding landfill and incineration may also reduce water, soil and air pollution generated by flue gas and leachate. Thus, biosludge conversion technologies aim at reducing the environmental impacts of disposal by increasing energy and nutrient recovery.

From an economic perspective, sludge treatment is one of the main cost factors of a P&P mill wastewater treatment process as it represents up to 50-60% of the total cost (Kinnunen et al., 2015). This is mainly due to the necessity and difficulty to dewater biosludge, as explained in section 2.2.1. Producing value-added products with biosludge can not only reduce environmental impacts of sludge handling but also contribute to mills' self-sufficiency in terms of fuel and create an additional revenue stream for the industry with soil

amendments. The development of alternative biosludge conversion methods is contributing to transforming pulp and paper mills into biorefinery, as part of circular economy objectives (Hämäläinen et al., 2022). As waste disposal regulations are getting tighter, this approach is becoming essential.

3. Biosludge conversion technologies

Biosludge conversion technologies can be categorized into three types of treatment, as shown on **Figure 4**: thermochemical treatment, biochemical conversion, and chemical conversion. Thermochemical treatment includes direct combustion, torrefaction, pyrolysis, gasification, hydrothermal carbonization. Biochemical treatment includes composting (also called aerobic digestion), fermentation and anaerobic digestion. Chemical conversion occurs through a process called transesterification (Banks, 2021; Praveen Kumar et al., 2020). This section first presents the different thermochemical treatment methods and then details the processes, challenges and end-products of combustion, hydrothermal carbonization, anaerobic digestion and fermentation.

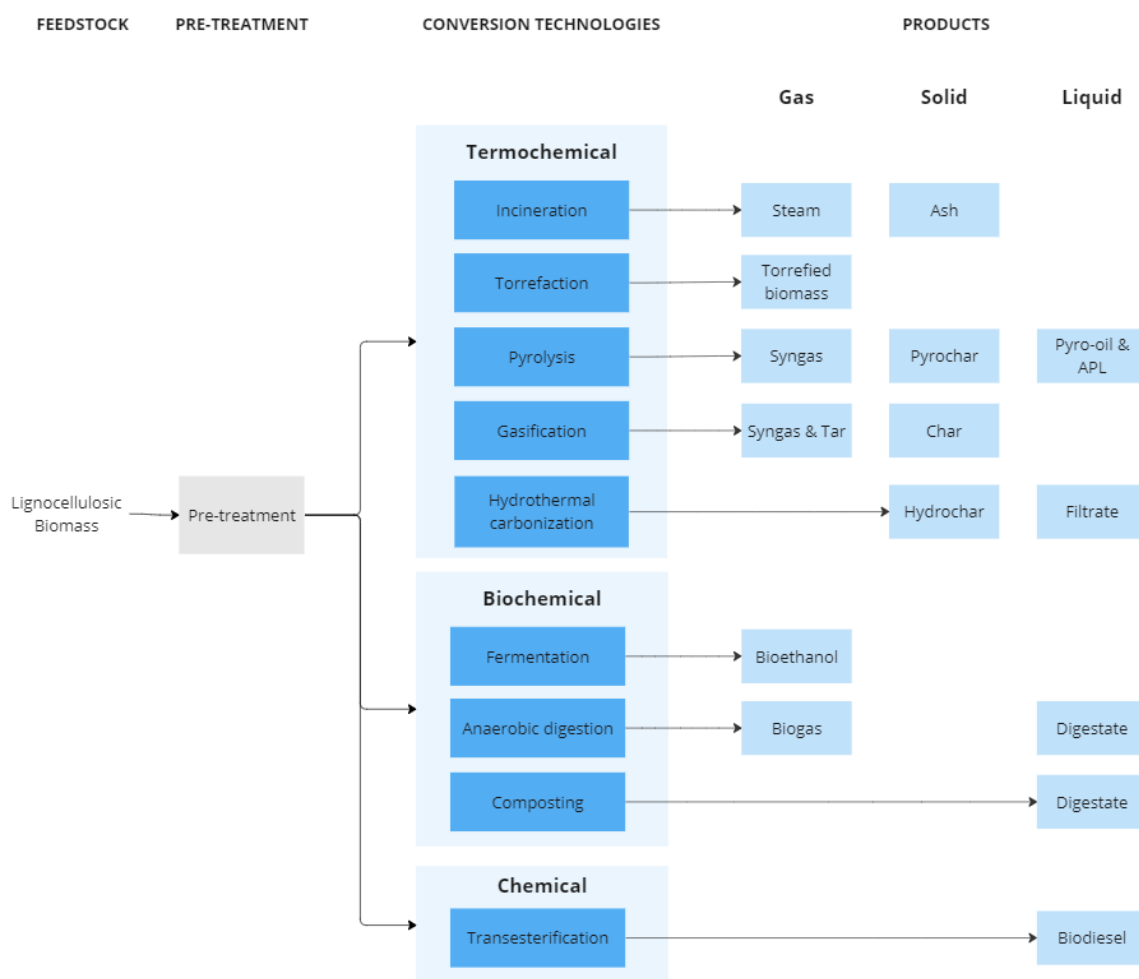


Figure 4. Biomass conversion technologies. APL: aqueous pyrolysis liquid (Adapted from: Ding et al., 2021; Pecchi & Baratieri, 2019).

3.1. Thermochemical conversion of biomass

During thermochemical conversion processes, biomass is heated at high temperature to be converted into energy and nutrients carriers. This process is gaining importance as it fits well the treatment of non easy-handled biomass, it also benefits from short process time and does not require maintenance of a biological community (Pecchi & Baratieri, 2019). Volume reduction is ensured, and energy recovery can be achieved as well. However, moisture content has to remain low (less than 30% for some reactions), to avoid excessive energy demand for drying. The different thermochemical treatment methods differ based on the environment in which the feedstock is converted, temperature range and heating rate,

influencing the end products generated. Table 3 compares the reaction conditions and products yields of different thermochemical reactions.

Table 3. Comparison of reaction conditions and typical product yields for thermochemical conversion processes (Adapted from: Libra et al., 2011; S. Meyer et al., 2011).

Process	Reaction conditions			Product distribution (wt%)		
	Medium	Temperature range [°C]	Typical residence time	Solid	Liquid	Gas
Torrefaction	Gaseous	200-300	10-60 min	61-84	n.a.	n.a.
Pyrolysis – slow	Gaseous	400	Hours-week	35	30	35
Pyrolysis – intermediate	Gaseous	500	10-20 s	20	50	30
Pyrolysis – fast	Gaseous	500	1 s	12	75	13
Gasification	Gaseous	600-1200	10-20 s	10	5	85
Hydrothermal carbonization	Aqueous	180-250	5 min-12 h	50-80	5-20	2-5
Hydrothermal liquefaction	Aqueous	300-400	10 s – 30 min	n.a.	Main product	n.a.
Hydrothermal gasification	Aqueous	350-750	10 s – 2 h	n.a.	n.a.	Main product

In the pyrolysis process, the reaction occurs in an oxygen-free environment and temperature ranges from 300 °C to 800 °C. Four main products are generated in this process: syngas, a combustible gas mainly containing CO, H₂, and CH₄; pyro-oil, an oily phase; aqueous pyrolysis liquid (APL), an aqueous phase; and pyrochar, a solid carbonaceous material. Ashes can also be produced in low quantities. Depending on the heating rate of the reaction and retention time, pyrolysis can be categorized into three sub-categories: slow, fast, and flash pyrolysis. The shorter the residence time and the higher the heating rate, the better the quality and yield of liquid products (Praveen Kumar et al., 2020). If the reaction occurs in a pressurized water environment, the reaction is called hydrothermal conversion or aqueous pyrolysis (Gollakota et al., 2018).

In gasification process, temperature ranges from 600 °C to 1200 °C and transformation takes place in a sub-stoichiometric oxygen environment. Biomass is converted into three products: syngas, a combustible gas mainly containing CO, H₂, and CH₄; tar, a condensable fraction composed of hydrocarbons; char, a solid carbonaceous material. Moisture content is suggested to be below 30% to ensure good quality products (Pecchi & Baratieri, 2019).

Torrefaction can be considered as a mild pyrolysis. The main objective of this process is to produce a solid product with better energy density and easily grindable to form pellets. Temperature ranges from 200 °C to 300 °C. Heating rate is quite slow to increase solid products yield (Acharya et al., 2015). Torrefaction is often considered as a pre-treatment as it transforms biomass for further use rather than direct use.

3.2. Direct incineration of biosludge

Incineration, or combustion, is the chemical process of burning biomass in the presence of air to transform chemical energy into heat (Praveen Kumar et al., 2020). Temperature ranges from 800 °C to 1000 °C. The moisture content of feedstocks is one of the limiting factors of incineration, the higher the water content, the lower the heating value as received. However, in the case of P&P mills biosludge, even after dewatering, the solid content remains around 8% to 11% (Saari et al., 2022). As sludge is incinerated with other wood-based waste such as fines and bark, total moisture content of feedstock is increased. The lower heating value as received (LHV_{ar}) of combustion feedstock is calculated using Equation 1. It takes into account the energy required to evaporate the water contained in the feedstock. For biosludge, when considering a lower heating value of the dry matter of 18.5 MJ/kg (Martinez et al., 2021) and moisture content of 8% after dewatering (Saari et al., 2022), the lower heating value as received of biosludge is negative and amount to -0.77 MJ/kg. When considering LHV_{ar} of 8.25 MJ/kg and 11.95 MJ/kg respectively for bark and fines, and a biomass boiler feedstock ratio biosludge/bark/fines of 22/71/7 (Saari et al., 2022), the average LHV_{ar} of the fuel is 6.5 MJ/kg. The combustion efficiency then varies from 70% to 90% according to (San José et al., 2014). In comparison, LHV_{ar} of sugarcane bagasse is 16.9 MJ/kg and LHV_{ar} of commercial coals used for the production of electricity ranges between 16.1 MJ/kg and 33.4 MJ/kg (Nunes et al., 2021). Direct combustion of biomass also results in the loss of nutrients contained in it (Kinnunen et al., 2015).

$$LHV_{ar} = LHV_{dry} \cdot (1 - MC) - l_{25} \cdot MC \quad (1)$$

Where:

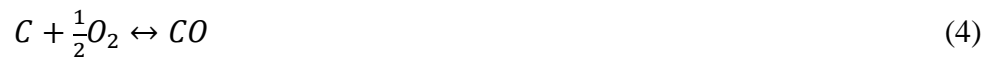
LHV_{ar} = Lower heating value as received [MJ/kg]

LHV_{dry} = Lower heating value of the dry matter [MJ/kg]

MC = Moisture content of feedstock

l_{25} = Heat of vaporization of water at 25°C [2.443 MJ/kg]

During combustion, two main waste streams are generated: flue gas and ash. Flue gas typically contributes to air pollution due to particulate matter and gaseous pollutants contained in it. When biomass is incinerated, elemental components of biomass are converted into hot gases (CO, CO₂, SO_x, NO_x) (Praveen Kumar et al., 2020). Major reactions occurring during combustion are given in Equations 2 to 8. Carbon monoxide (CO) and carbon dioxide (CO₂) are generated during the combustion of a fuel, as the result of the reaction between carbon content in fuel and oxygen (Eq. 3 & 4). If the combustion is complete, carbon dioxide is produced, otherwise carbon monoxide is formed instead. CO₂ is a greenhouse gas that contributes to global warming and CO is dangerous for human health (Kennes & Veiga, 2013).



The U.S. Environmental Protection Agency defines two types of CO₂ emissions: biogenic CO₂ emissions that results from the combustion, decomposition or processing of biomass, and fossil CO₂ emissions resulting from the processing of fossil fuels, peat, and mineral sources of carbon (U.S. Environmental Protection Agency, 2011). This concept can be extended to other carbon-based greenhouse gases (GHG) such as methane and carbon monoxide. The distinction between biogenic and fossil emissions is based on the wide difference in time scales between carbon circulation resulting from anthropogenic biomass processing and fossil fuel processing. In the first case, carbon sources are originated from plants or trees that grew over the scale of months to decades, during which they removed carbon from the atmosphere. The carbon absorbed is then released back to the atmosphere during combustion process. Therefore, biogenic CO₂ emissions will not inevitably increase the amount of CO₂ in the atmosphere over policy-relevant time scales. On the other hand, fossil fuels have formed over thousands of years and the carbon contained in them was not part of the active carbon cycle anymore. Their combustion thus releases historical carbon to the atmosphere and increase the

concentration of CO₂ on policy-relevant timeframes (U.S. Environmental Protection Agency, 2011). Considering the short timeframe of carbon circulation in the case of biomass processing, the majority of bioenergy policies assumes that CO₂ emissions from renewable biomass processing are offset by CO₂ sequestration in growing biomass, therefore bioenergy is considered climate neutral (Cherubini et al., 2012). The 2009/28/EC Directive specifies that GHG emissions resulting from the use of biofuels and bioliquids shall be taken to be zero. In the case of pulp and paper biosludge, its combustion to produce heat and electricity results in biogenic emissions that should not be taken into account according to the 2009/28/EC Directive. However, the scientific and policy community is facing an increasing debate on whether to include biogenic emissions into environmental impact studies (Cherubini et al., 2012). The assumption of carbon neutrality for bioenergy raises a temporality issue. It implies that CO₂ emitted into the atmosphere is immediately re-absorbed by photosynthesis. Yet, the global warming potential of GHG depends on their residence time in the atmosphere, the delay between emission and absorption of biogenic GHG thus has an impact on the environment (Pelletier et al., 2019). (Cherubini et al., 2012) analysed in more detail the impact of biogenic GHG emissions on global warming potential. Two important observations have been made: (1) GHG emissions resulting from the processing of slow growing biomass species, like forest wood, stay longer in the atmosphere and thus have a non-negligible impact on global warming; (2) changes in albedo occurring after biomass harvest cause a cooling contribution, especially in region affected by seasonal snow cover because of the higher reflective property of snow. Those observations show the complexity of characterising the global warming potential of biogenic carbon emissions, as they can either contribute to warming or cooling the atmosphere, depending on the biomass species, local climate conditions, and time horizons. Several studies have dealt with the influence of biogenic carbon impact factor on environmental impact assessments of biomass-to-heat systems (Cherubini et al., 2012; Nian, 2016; Pelletier et al., 2019), and some authors also computed site and species-specific biogenic carbon equivalency factors (BCEF) (Cherubini et al., 2012).

Another source of air pollution during combustion is nitrogen oxides. The major pollutants of this group are nitric oxide (NO) and nitrogen dioxide (NO₂), referred as NO_x. They are produced by the reaction between nitrogen present in fuel or atmosphere and oxygen, which significantly occurs at high temperature. NO is first generated during combustion and part of it is then converted to NO₂ (Eq. 5 & 6). Sulphur oxides (SO_x) include sulphur dioxide (SO₂) and sulphur trioxide (SO₃). They are generated by the reaction between sulphur present in fuel and oxygen (Eq. 7 & 8). SO₂ is primarily formed during combustion and up to 5% of it is then converted to SO₃, a more corrosive and toxic component. This additional reaction mainly occurs at higher temperature and in the presence of excess oxygen. Nitrogen and sulphur oxides contribute to global warming, may lead to the formation of acid rains, and can cause human respiratory issues. (Kennes & Veiga, 2013.)



The solid waste stream of combustion is ash, which is mainly formed by inorganic elements. As detailed in part 2.2.3, they have negative impact on combustion process and can be challenging to handle. Commonly, ash is landfilled (Mohammadi et al., 2019b).

Incineration of biosludge presents several drawbacks. The main disadvantage is the negative lower heating value of biosludge, due to its high moisture content, compared to bark and fines heating values, which results in a loss of steam and power generation (Saari et al., 2022). Besides, operational problems related to biosludge incineration include ash fusion, scaling, high emissions levels of SO₂ and NO_x, difficulties with boiler feeding and decreased bed temperatures (Stoica et al., 2009). Thus, there is a need for alternative options. By using biochemical and thermochemical treatment processes, value can be added to biosludge in regard of energy and nutrient contents (Hämäläinen et al., 2022).

3.3. Hydrothermal carbonization (HTC)

Hydrothermal carbonization is a thermochemical process that converts wet lignocellulosic biomass into a stable coal-like product called hydrochar. HTC can also be called hydrothermal pre-treatment, wet torrefaction, or wet pyrolysis (Libra et al., 2011; Mäki, Saastamoinen, Melin, Matschegg, & Pihkola, 2021). This section details the process and end-products characteristics of HTC.

Even if industrial scaling of hydrothermal carbonization of biosludge has been proven feasible, this technology is not yet widely developed in pulp and paper industry. As of today, only one industrial-scale demonstration is on-going, at Stora Enso's Heinola mill in Finland (Mäki, Saastamoinen, Melin, Matschegg, & Pihkola, 2021). The development of HTC in P&P industry is facing different challenges: hydrochar produced from the process is still declared as waste in Finland, so End-of-Waste procedure is required to convert it to a marketable resource (Mäki, Saastamoinen, Melin, Matschegg, Davidis, et al., 2021). Hydrochar also needs standardization to facilitate its integration on the market. The conversion technology lacks readiness, compared to other biosludge handling options such as anaerobic digestion. Long-term studies are thus necessary to analyse the environmental impacts of the process and characteristics of the end-products. Heinola's project is aiming at filling the gaps on HTC implementation in P&P industry. From an economic point of view, investment, operation, and maintenance costs are barriers to HTC development. To face this issue, hydrochar has to be sold at an acceptable price and close to the mill to reduce transportation costs. (Mäki, Saastamoinen, Melin, Matschegg, Davidis, et al., 2021) estimated that hydrochar market price needs to reach at least 58 €/MWh (instead of 40 €/MWh in 2021), to achieve economic feasibility of HTC integration into Nordic P&P mills.

3.3.1. Process

During hydrothermal carbonization, the organic matter of biomass is thermochemically decomposed in subcritical liquid water (Libra et al., 2011). The reaction occurs in a high-pressure reactor that allows pressure to rise so that the feedstock stays in a liquid state.

Temperature ranges from 180 °C to 250 °C and pressure goes up to 20 bars. The main product of the reaction is hydrochar, a solid material that stands for 50-90% of initial feedstock dry weight. Liquid is also generated in lesser amount (5-20% of feedstock dry weight) and is called HTC liquor or filtrate. Very little gas is produced (less than 5%). Reaction time is usually from 5 minutes to 12 hours (Saari et al., 2022). This process fits well wet biomass treatment, such as biosludge, as it does not require excessive energy for drying before or during the process.

Several reaction mechanisms occur during HTC, including hydrolysis, condensation, decarboxylation, and dehydration reaction (Tasca et al., 2019). First, cellulose, hemicellulose and lignin contained in biomass are converted into monosaccharides and disaccharides through hydrolysis and dehydration. Those intermediate products are then dehydrated, hydrolysed and decarboxylated. Finally, recondensation of the newly formed compounds leads to the formation of hydrochar.

3.3.2. Hydrochar properties and applications

(Martinez et al., 2021) analysed the properties of hydrochar produced from hydrothermal carbonization of pulp mill biological sludge and the influence of temperature on the reaction (from 180 °C to 240 °C). Many chemical and physical changes occur during HTC. First, hydrochar typically weight 50% to 80% of initial feedstock weight on a dry basis and 80% to 95% of its energy content. Volume reduction and energy densification are thus achieved during HTC. The process also increases hydrophobicity of biosludge. This characteristic is emphasised with reaction severity. Moisture content is consequently reduced in the process. For biosludge with initial moisture content around 92%, HTC can produce hydrochar with less than 7% moisture content. Higher hydrophobicity and lower moisture content make biomass easier to dry, store and transport. In HTC process, volatile matter is reduced up to 16 wt% and fixed carbon is increased up to 14 wt%. Ash content is barely changing, with a maximum reduction of 3.82 wt% at lower temperature (180 °C). Hydrochar also benefits from a high porosity which is suitable for its use as activated carbon or soil conditioner (Mannarino et al., 2022).

When comparing the elemental composition of biosludge and hydrochar, carbon content is increased during HTC, up to 7.81 wt% (Martinez et al., 2021). Hydrogen content stays quite constant and oxygen content is decreased, up to 8.43%. Those changes intensify with reaction severity. Biosludge undergoes a coalification process during HTC. The degree of coalification, which is of high interest in biomass thermochemical conversions, can be expressed on the van Krevelen diagram, as shown on **Figure 5**. It presents the H/C-ratio and O/C-ratio of different materials to analyse their calorific value potential (Basu, 2018). During HTC, H/C-ratio and O/C-ratio are decreasing with reaction severity. The elemental composition of hydrochar thus more closely resemble the one of coal. Those modifications have a positive impact on hydrochar higher heating value, a 25% increase can be reached after HTC of biosludge at 240 °C.

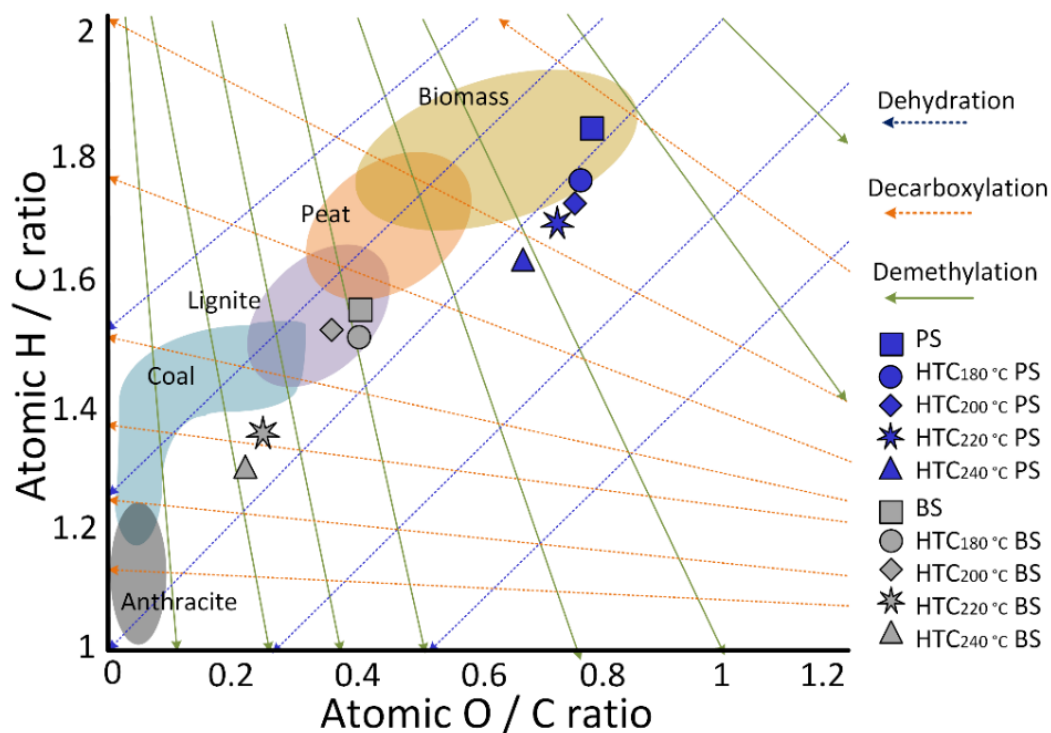


Figure 5. Development of the elemental composition of biomass during HTC. PS: Primary sludge; BS: Biosludge. The tendency of the dehydration, decarboxylation and demethylation reactions is shown by the arrows. (Martinez et al., 2021)

Hydrochar potential for energy recovery has been analysed in different studies (Hämäläinen et al., 2021, 2022; Kim et al., 2014; Mannarino et al., 2022; Saari et al., 2022), only a few of them focus on hydrochar derived from pulp and paper mills' biosludge. (Martinez et al., 2021) reveals that HTC improve biosludge characteristics as a fuel because of enhanced

energy efficiency and lower corrosive components contents. Higher heating value of hydrochar has been proven to be higher than for biosludge. Energy densification increases with reaction severity and can reach 1.25 at 240 °C. Chlorine, sodium, and potassium contents reduction implies less ash-related problems during hydrochar combustion. Nitrogen and sulphur contents are also lower which reduces the environmental impacts of combustion. Hydrochar can be used as a fuel on the mill, to enhance energy self-sufficiency, or can be sold, to provide an additional revenue.

Hydrochar utilization as a fertilizer is also promising because of the significant reduction of heavy metals concentration during HTC (Martinez et al., 2021). (Rillig et al., 2010) analysed the effects of hydrochar derived from beet root chips on plant growth. The ultimate composition of the hydrochar used in the study is very similar than the one of the hydrochar produced by HTC reaction of P&P biosludge at 240 °C in (Martinez et al., 2021). C content is 53.2% instead of 52.57%, H content is 5.3% for 5.7% in biosludge-derived hydrochar and N content is 2.3% instead of 3.0%. (Rillig et al., 2010) noticed a deleterious effect of hydrochar on plant growth when used at high addition rate. Thus, the authors advise a cautious approach in using hydrochar as a soil additive. The residence time of hydrochar in soil also seems to be shorter than typical biochar produced from pyrolysis.

3.3.3. Filtrate properties and applications

While the use of hydrochar for energy and nutrient recovery has been the main focus of HTC research in the last decades, HTC filtrate properties and applications is lacking attention (Langone & Basso, 2020). The chemical properties of HTC filtrate can make its handling challenging. Different research groups have analysed the impacts of HTC filtrate utilization in various applications. The first option is to dispose of the filtrate into the WWTP of the mill, for further treatment. Several studies have been conducted to determine the impacts of filtrate recirculation into the HTC reactor. This has multiple advantages: it regulates the moisture content in HTC reactor, enhances dewaterability, increases mass and energy yields of hydrochar (Banks, 2021; Langone & Basso, 2020). Due to the hydrolysed sugars, high organic content, and volatile fatty acids contained in it, HTC filtrate use for anaerobic digestion is also promising (Mannarino et al., 2022).

3.4. Anaerobic digestion (AD)

Anaerobic digestion is the microbially mediated degradation of complex organic matter into simple organic compounds and dissolved nutrients under anoxic conditions (Kennes & Veiga, 2013; Nikolausz & Kretzschmar, 2021). The main product of this process is a biogas mainly containing methane (CH₄) and carbon dioxide (CO₂). Digestate, which can be utilized as a biofertilizer, is also produced during anaerobic digestion. Anaerobic digestion can be engineered or can also happen naturally when biomass is degraded by bacteria in an environment with little or no oxygen.

Due to its energy and nutrient recovery potential from waste biomass, anaerobic digestion has been attracting increasing industrial interest since the middle of the 20th century, with a focus on methane production from agricultural residues. Even though AD is a mature biochemical technology, research is still needed due to the diversity of feedstock as well as products that can be generated from the process. In 2018, the installed capacity worldwide of industrial biogas plants reached 19 GW and 6,586 TWh of electrical power were generated. Most of the biogas plants are located in Asia (40%), Europe (20%) and North America (19%) (Nikolausz & Kretzschmar, 2021). The recent fast development of AD (mostly helped by diverse subsidy systems), the diversity of feedstock that can be used, and the flexibility of energy generation make AD a major player in the energy transition. However, AD technologies have to face different challenges, such as the limited availability of feedstock, the competitive use of biomass with food and feed production, the reduction of subsidies, and the low competitiveness of products. Its use is thus most fitted for biomass waste upgrading. Despite being widely used to treat agricultural residues, AD has only recently started to be used for municipal wastewater and is not much applied to pulp and paper biosludge (Kinnunen et al., 2015).

3.4.1. Process

Anaerobic digestion of organic material can be divided into four steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis, as shown on Figure 6 (Kennes & Veiga, 2013). In the first step, that is hydrolysis, organic matters containing complex compounds

are broken down, to become available for the next steps. During acidogenesis, also called fermentation, sugars, amino acids, and long-chain fatty acids are converted into CO₂, ammonia, hydrogen, alcohols and volatile fatty acids. In acetogenesis, the previously formed compounds are converted into acetic acid, carbon dioxide and hydrogen. In the final step, that is methanogenesis, the latter products are converted into CH₄ and CO₂.

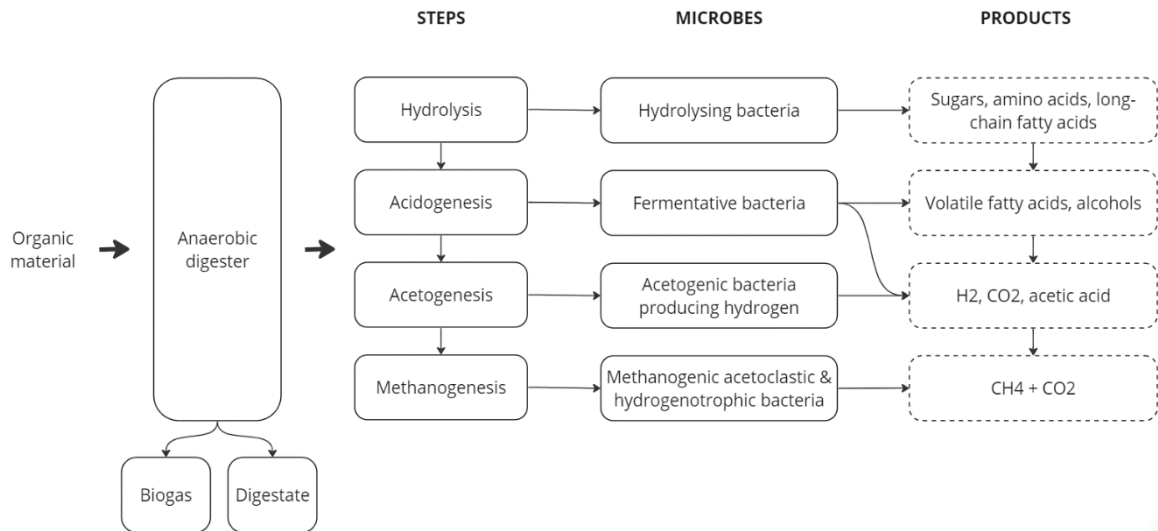


Figure 6. Schematic representation of anaerobic digestion process (Adapted from: Kennes & Veiga, 2013; Praveen Kumar et al., 2020; Veluchamy & Kalamdhad, 2017).

Despite its double recovery potential, anaerobic digestion is facing several technical challenges. First, the operation of the process can be unstable due to the nature of the reactions and environmental conditions (temperature, pH, concentrations of some substances in the feedstock). The maintenance of the microbial community inside the reactor can be quite complex. Then, digestate can be of low quality due to its poor retention capacity for nutrients. (Pecchi & Baratieri, 2019). One condition for the feasibility of anaerobic digestion is that COD concentration in feedstock has to be relatively high (> 1 g/L) (Meyer et al., 2014). The challenges related to P&P mill biosludge anaerobic digestion are influenced by the type of raw materials, pulping processes and wastewater treatment processes (Mäki, Saastamoinen, Melin, Matschegg, & Pihkola, 2021). Several studies have pointed out the low degradability of pulp and paper biosludge because of its high lignocellulosic content (T. Meyer & Edwards, 2014). This results in a slow or incomplete hydrolysis, which implies longer retention times, larger reactors, and thus higher investment costs. **Table 4** details the difference between different feedstock in terms of lignocellulosic content. Section 3.5.1

gives a more detailed explanation of the lignocellulosic complex. Those aforementioned challenges explain the low implementation of anaerobic treatment in pulp and paper mills.

Table 4. Lignocellulosic content of different feedstocks.

Parameter	Pulp & paper biosludge (Meyer et al., 2014)	Municipal biosludge (Meyer et al., 2014)	Bagasse (Aggarangsi, 2023)	Wheat straw (Aggarangsi, 2023)	Poultry litter (Zhongqi, 2012)
Cellulose [% DS]	19-27	~1	30-50	30	14.3
Lignin [% DS]	36-50	<0.1	20-35	15	1.8

Anaerobic digestion process is influenced by several factors. Temperature has a great influence on AD, not only from a kinetic point of view but also on the thermodynamic of the reactions (Kennes & Veiga, 2013). Two ranges of temperature can be chosen: a mesophilic anaerobic digestion occurs between 30 °C and 35 °C and a thermophilic reaction occurs from 50 °C to 60 °C. Even if the latter is faster, it is also more unstable and thus more complex to control. Mesophilic AD is therefore often preferred. Temperature stability during AD is also very important. Methanogenesis is the most sensible step to temperature variations. If temperature changes more than 1° C/d, the process might fail, because of methane-forming bacteria inhibition.

Methanogenic bacteria are also very sensitive to the concentration of some substances that can inhibit their activity and cause instability of the process. Anaerobic digestion can face two different types of inhibition (Pecchi & Baratieri, 2019). Primary inhibition results from the high concentration of some substances, called inhibitors, in the feedstock itself. The main known inhibitors are free ammonia, hydrogen sulphides, salinity, xenobiotic compounds (Kennes & Veiga, 2013). However, no threshold limit exists because their influence on methanogenic bacteria depends on different factors such as temperature, pH, concentrations of other substances. Some of them can even have a positive impact on anaerobic digestion at low concentration. Secondary inhibition results from the excessive production of reaction intermediates, such as VFAs, that causes acidification of the organic material. Methanogenic bacteria are very sensitive to pH and perform best within a pH range between 6.8 and 7.2. However, when anaerobic reactions occur, the intermediate compounds generated lower the

alkalinity of the material, thus inhibiting methanogenic bacteria activity. Only feedstocks containing low concentrations of inhibitory or toxic compounds are suitable for anaerobic digestion.

Different parameters are used to characterise an anaerobic digestion process. First, the hydraulic retention time (HRT) is a measure of the time the feedstock liquid (substrate) spends in the reactor (Kennes & Veiga, 2013). It is defined as the ratio between the volume of the reactor and the substrate flow rate (Equation 9). HRT usually varies from 15 to 30 days in a mesophilic digester, and from 12 to 14 days for a thermophilic digester. The value is influenced by the feedstock composition and reaction temperature. If HRT is too low, the organisms don't have enough time to completely degrade the organic matter. If HRT is too high, overload of the system might occur.

$$HRT = \frac{V}{Q} \quad (9)$$

Where:

HRT = hydraulic retention rate [d]

V = reactor volume [m³]

Q = flow rate [m³/d]

Solids retention time (SRT) is measuring the digestate age (Kennes & Veiga, 2013). It is defined as in Equation 10. A high SRT increases biogas production by favouring methanogenesis. While HRT needs to be quite short to treat large volumes of sludge quickly and economically, SRT needs to be maximized for higher biogas production. HRT and SRT are often the same, but some AD reactors enable to decouple SRT and HRT to achieve better performances (T. Meyer & Edwards, 2014).

$$SRT = \frac{VX}{QX_e} = HRT \frac{X}{X_e} \quad (10)$$

Where:

SRT = solid retention rate [d]

X = concentration of microorganisms inside the digester [kg.m⁻³]

X_e = concentration of microorganisms in the effluent stream [kg.m⁻³]

Organic load rate (OLR) is defined as the amount of substrate feed into the digester volume in a given amount of time (Equation 11) (Kennes & Veiga, 2013).

$$OLR = \frac{QS}{V} = \frac{S}{HRT} \quad (11)$$

Where:

OLR = organic load rate [kg/(m³.d)]

S = substrate concentration [kg/m³]

Anaerobic digestion reactors, also called biogas plants, are classified into two categories: batch type or continuous type (Praveen Kumar et al., 2020). In case of a batch type plant, feedstock is charged at 50-60 days intervals, until the digestion process is completed. For continuous type plants, a certain quantity of biomass is input daily. Continuous type plants can be based on constant pressure or constant volume. The most common anaerobic systems used for P&P industry's sludge are up-flow anaerobic sludge blanket (UASB) and internal circulation reactor. They both makes it possible to handle large volumetric flows and high chemical oxygen demand (COD) loads (Mäki, Saastamoinen, Melin, Matschegg, & Pihkola, 2021).

3.4.2. Biogas production

Biogas mainly contains CH₄ and CO₂, typical proportions are respectively 60% and 40%. Other compounds found in minor proportions are H₂, H₂S, CO, siloxanes, mercaptans (Kennes & Veiga, 2013). The energy conversion efficiency of anaerobic digestion is 60% to 90%, which is the energy available from the combustion of biogas as a percentage of the feedstock dry matter heat of combustion (Praveen Kumar et al., 2020). To evaluate the methane production potential of a substrate, the biochemical methane potential (BMP) is usually used. It can be expressed in two ways: the ratio between the volume of biomethane produced and the VS content in the feedstock, or the ratio between the volume of biomethane produced and the amount of COD removed. BMP is measured during laboratory tests (Feiz et al., 2022). Concerning biosludge from pulp and paper industry, the methane potential is rather low compared to other usual feedstocks, with a BMP widely varying from 30 to 200 L CH₄/kg VS fed (T. Meyer & Edwards, 2014) and rarely exceeding 100 L CH₄/kg VS fed

(Karlsson et al., 2011; Kinnunen et al., 2015). (Kinnunen et al., 2015) analyzed the biomethane potential of P&P mill biosludge. They revealed methane yields of 70-78 L CH₄/kg VS for HRTs of 20 and 14 days. In comparison, BMP of household food waste is around 400 L CH₄/kg VS (Feiz et al., 2022) and BMP for manure varies from 200 to 550 L CH₄/kg VS fed (Aggarangsi, 2023). As BMP is measured under laboratory environment, the specific methane production (SMP) is lower than the BMP and accounts for about 90% of the BMP under well-managed anaerobic digestion configuration (Feiz et al., 2022).

Different outcomes are possible for biogas valorisation in P&P industry. Biogas can be directly used at the mill to replace fossil fuels. It increased the energy self-sufficiency of the mills and replace the use of fossil fuels. Biogas can also be sold to external markets. Golbey French P&P mill and Saugsbrugs Norwegian mill are respectively selling to the public gas distribution system and a heavy vehicles gas supplier. This strategy gives an additional revenue stream to the mills. Another way to use biogas is to transform it into liquefied biogas (LBG) and then sell it for later use as a fuel. Stora Enso's mill in Nymölla is applying this valorisation path. (Mäki, Saastamoinen, Melin, Matschegg, & Pihkola, 2021).

Research on AD has studied the potential benefits of pre-treatment of feedstock on methane yield. (Kinnunen et al., 2015) analysed the impact of thermal pretreatment of P&P biosludge on methane production. When biosludge is warmed up for 20 min at 105-134 °C, biomethane potential increases with temperature by up to 140% compared to untreated biosludge, during the first 10 days of AD. It has been noticed that methane production is faster in those conditions, most of the additional methane is produced during the first 10 days. Shorter HRT is thus achievable, resulting in smaller reactor size and reduced energy consumption. Several recent studies have focused on hydrothermal pre-treatment of biomass for anaerobic digestion. According to (Ahmad et al., 2018), HTC pre-treatment at relatively low temperature (< 180 °C) can increase methane yield by 58%, while pre-treatment at more than 200 °C results in lower degradability and methane yield.

3.4.3. Digestate properties and applications

Digestate is characterised by a high moisture content, typically higher than 90% (Langone & Basso, 2020). The properties and composition of digestate depends on the feedstock used for anaerobic digestion and the type of biogas reactor. In the case of digestate produced from P&P secondary sludge, moisture content is around 91 wt% and carbon content about 0.36 kg C / kg DS (Eikelboom et al., 2018). Digestate is usually dewatered to ease its handling.

Digestate can be directly disposed to landfill or used for land application, but those disposal methods result in concerning environmental and health impacts. (Eikelboom et al., 2018) compared different alternative recycling methods for anaerobically digested kraft pulp mill sludge, using a multi-criteria decision analysis. When considering environmental, economic, and technical impacts of each option, it has been found that the best disposal method is composting, followed by incineration, land application, pyrolysis/gasification, algae production. The worst scenario is landfill disposal. Composting produces a low-cost fertilizer for agriculture, its environmental impacts are reduced, and the process is financially and technically easy to implement. (Mohammadi et al., 2019b) found out that drying and pyrolysis of sludge digestate is a better environmental option than incineration when considering the life cycle impacts of both disposal methods. However, its implementation in P&P mills is much more challenging and requires high-cost investments.

Several studies have also analysed the hydrothermal carbonization of digestate of sewage sludge (Langone & Basso, 2020). Digestate can be treated directly by HTC or be dewatered before. Coupling both treatment methods, with process waters recycling in the biogas plant, can increase energy recovery from biosludge and improve the properties and yield of the products.

3.5. Fermentation

Fermentation is the biochemical conversion of sugars contained in lignocellulosic biomass into ethanol by micro-organisms.

3.5.1. Process

The most commonly used method to convert lignocellulosic biomass into bioethanol is via enzymatic hydrolysis and fermentation, see Figure 7. This biochemical process relies on the properties of the lignocellulosic complex of the feedstock. The lignocellulosic complex is formed by three polymers: lignin, hemicellulose and cellulose. In this cell-wall constituent, cellulose is the main structural component. Hemicellulose is forming a layer around cellulose fibres and lignin acts as a glue between them (Dutta et al., 2022). Ethanol is produced from polysaccharides contained in cellulose. The higher the content of cellulose and hemicellulose in biomass, the better the yield. On the opposite, lignin content has to be low for a better ethanol production. In biosludge from pulp and paper mills, cellulose share of total solids varies from 19% to 27% and lignin share varies from 36% to 50% (T. Meyer & Edwards, 2014), while the composition of typical lignocellulosic raw materials for fermentation is rather between 31% and 42% of cellulose and 17% to 26% of lignin (Kennes & Veiga, 2013). Ethanol production from pulp and paper biosludge can vary from 149 to 197 kg of ethanol per ton of dry biosludge (Chen et al., 2014).

Before hydrolysis and fermentation can happen, it is necessary to break down the lignocellulosic complex through pre-treatment to give access to the polysaccharides contained in cellulose and hemicellulose. According to (Boshoff et al., 2016), the advantage of pulp and paper mills sludge is that the lignocellulosic complex has already been broken during the paper making process, which makes it possible to avoid the energy intensive and costly pretreatment.

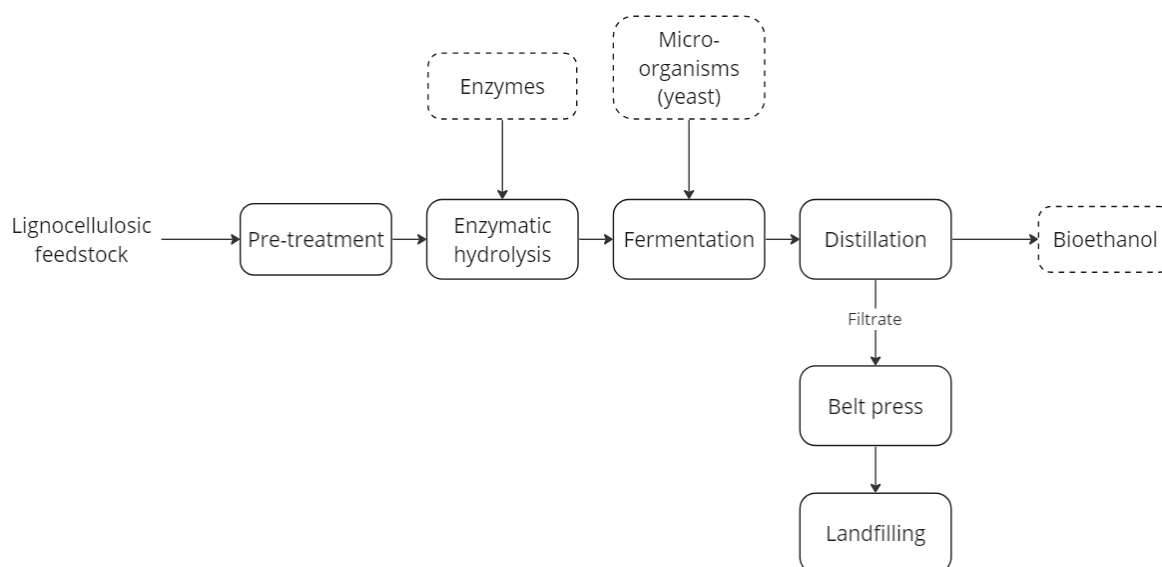


Figure 7. Process overview of lignocellulosic bioethanol production via enzymatic hydrolysis and fermentation (Adapted from Gilpin and Andrea 2017).

Once the lignocellulosic complex is broken, cellulose and hemicellulose are hydrolysed into fermentable sugars. Hydrolysis can be carried out chemically or enzymatically, with the second method being the most common one. For the production of bioethanol from lignocellulosic biomass, the enzyme typically used is cellulase. It comprises endoglucanases, exoglucanases, and β -glucoside. When ethanol is produced from starch, like corn starch, the production process rather uses α -amylase and glucoamylase enzymes (Dunn et al., 2012). Fermentable sugars are then converted into ethanol by microorganisms during the fermentation process (Kennes & Veiga, 2013). The most common microorganism is yeast, typically *Saccharomyces cerevisiae* (Dunn et al., 2012).

Commonly, enzymatic hydrolysis is carried out in a separate reactor and then fermentation occurs in a second reactor, which is called separate hydrolysis and fermentation (SHF). However, the two processes can also be combined in one reactor, which is called simultaneous saccharification and fermentation (SSF). While this second technology benefits from lower product inhibition of the enzymes, it is necessary to compromise on the reaction conditions between the optimal conditions for hydrolysis and those for fermentation (Kennes & Veiga, 2013). SHF is thus easier to handle and allows more system flexibility (Sebastião et al., 2016).

Enzymatic hydrolysis requires large quantity of cellulase enzyme (CE). Even if the production of CE is often excluded from the LCA studies of lignocellulosic bioethanol production, several studies have demonstrated that this process contributes highly to the environmental and economic life cycle impacts of bioethanol production (Gilpin & Andrae, 2017). However, cellulase enzyme technology is quickly evolving, thus reducing the need for enzyme in bioethanol production. Cellulase dosage is therefore subject to high variations and LCA studies of lignocellulosic bioethanol production requires frequent reevaluation (Dunn et al., 2012). Based on data available in literature, cellulase dosage for P&P biosludge SHF was calculated, the required dosage can vary from 9 kg/t dry fed to 100 kg/t dry fed, with a mean value of 33,09 kg/t dry fed and standard variation of 135% (Chen et al., 2014; Dunn et al., 2012; MacLean & Spatari, 2009). (Gilpin & Andrae, 2017) evaluated the environmental impact of cellulase production in six impact categories using CML impact method, as well as the cumulative energy demand of the process. The study estimated a global warming potential (GWP100) of 14,7 kg CO₂ eq. and a cumulative energy demand of 120.2 MJ to produce one kilogram of cellulase. Therefore, for the hydrolysis of 1 ton dry of P&P biosludge, global warming potential would account for approximately 486 kg CO₂ eq. and cumulative energy demand would reach 4,0 GJ.

3.5.2. Application

Bioethanol is a renewable fuel that can replace fossil fuels, especially gasoline in the transport sector (M. Wang et al., 2012). Two different types of bioethanol are currently produced. The most common type is the conventional first-generation bioethanol that uses energy crops to produce biofuel. This production method is often competing with arable land for food and feed. Second-generation biofuels, on the opposite, are produced from biomass waste and residues, such as biosludge (Sebastião et al., 2016). The revised Renewable Energy Directive (EU) 2018/2001 (RED II) sets the goal to reach 14% of renewable energy in the transport sector mix of each European Union Member State by 2030, with a minimum share of 3.5% of second-generation biofuels. Second-generation biofuels are thus expected to play an important role in the energy transition of the transport sector.

4. Life cycle assessment of pulp and paper mill biosludge treatment

This LCA study was conducted according to the methodology given by the International Organisation for Standardisation in SFS-EN ISO 14040:2006 and SFS-EN ISO 14044:2006. The four-step process includes: goal and scope definition, inventory analysis, impact assessment and result interpretation. This section details the outline of the study, the different biosludge treatment systems analysed, the calculation of the emissions and the environmental impacts associated.

4.1. Goal and scope definition

The objective of this study is to analyse the environmental impact of different energy and nutrient recovery methods from biosludge in pulp and paper industry. This research aims to guide the implementation of circular economy into this industry by maximising resource efficiency through waste recovery. The present LCA study considers the biosludge produced from secondary treatment at pulp and paper wastewater treatment plants. The system boundaries comprise all processes from biosludge collection and pretreatment to conversion processes and final disposal. Five alternative scenarios are analysed in a comparative way: (S0) direct incineration with energy recovery; (S1) HTC and energy recovery of hydrochar through incineration; (S2) HTC and nutrient recovery of hydrochar as a soil amendment; (S3) AD of biosludge followed by HTC of digestate and use of hydrochar as a soil amendment; (S4) separate hydrolysis and fermentation of biosludge. S0 is used as a baseline scenario, as incineration is the most common treatment method in Finland. According to previous LCA studies of sludge treatment systems, the functional unit (FU) commonly used is mass of feedstock or mass of energy generated. In this study, the FU selected is the treatment of 1 tonne of dry matter biosludge, considering the different properties of by-products in each scenario (Mohammadi et al., 2019a, 2019b; Sebastião et al., 2016). System boundaries of each system are presented in Figure 8. System expansion was used to account for the use of co-products from each system as energy or nutrients carriers. System expansion is detailed in section 4.2.7.

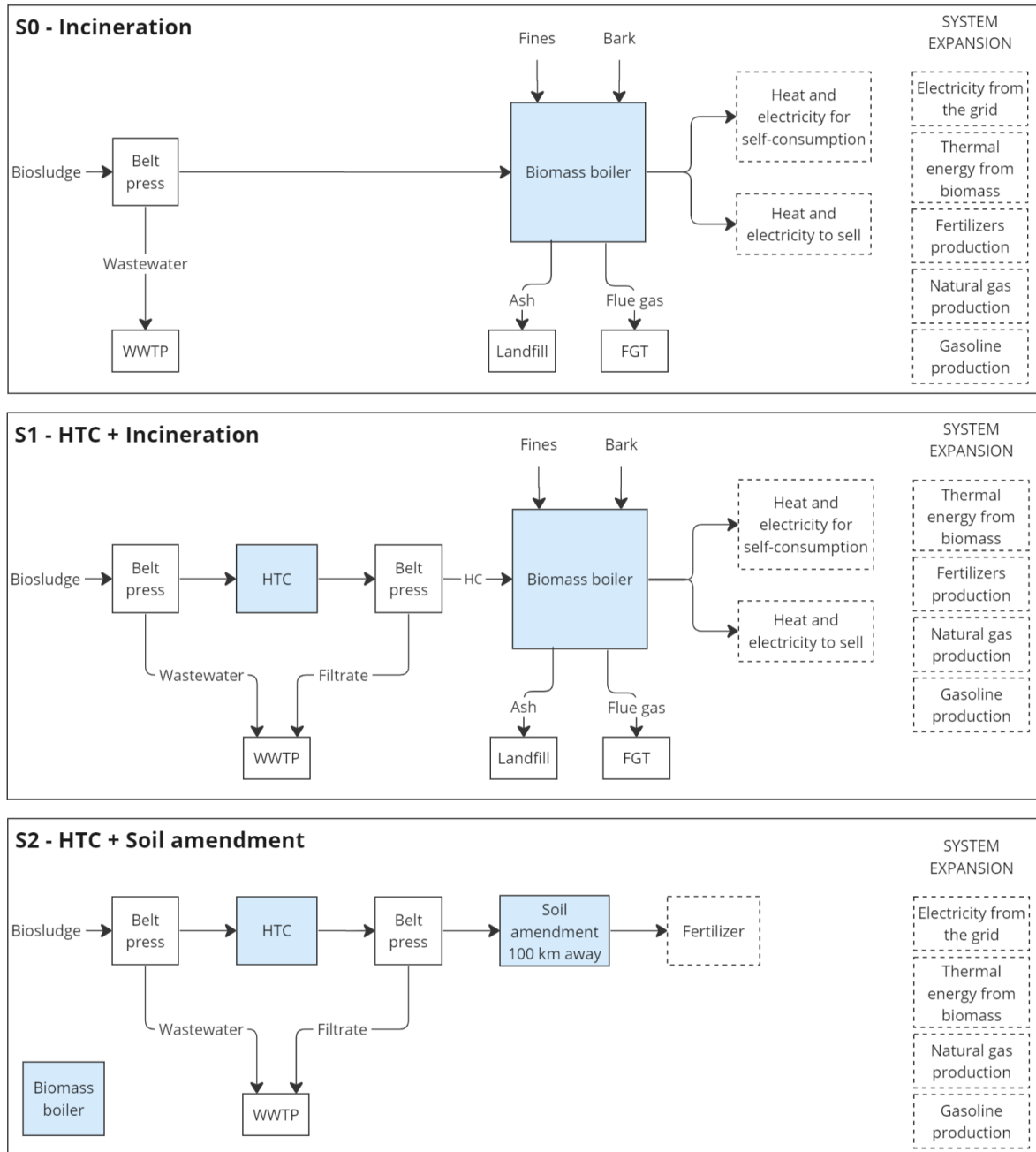


Figure 8. Life cycle diagrams for System 0 (S0 - Incineration), System 1 (S1 - HTC + incineration), System 2 (S2 - HTC + soil amendment), System 3 (S3 - AD + HTC + soil amendment), System 4 (S4 - Separate hydrolysis and fermentation). The life cycle diagram for the boiler in S2, S3, S4 is represented separately. AD: anaerobic digestion; FGT: flue gas treatment; HC: hydrochar; WWTP: wastewater treatment plant.

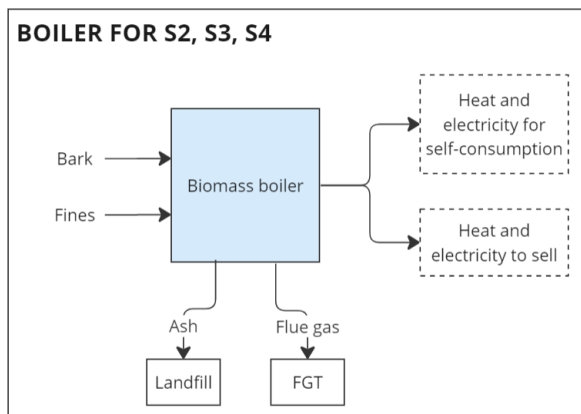
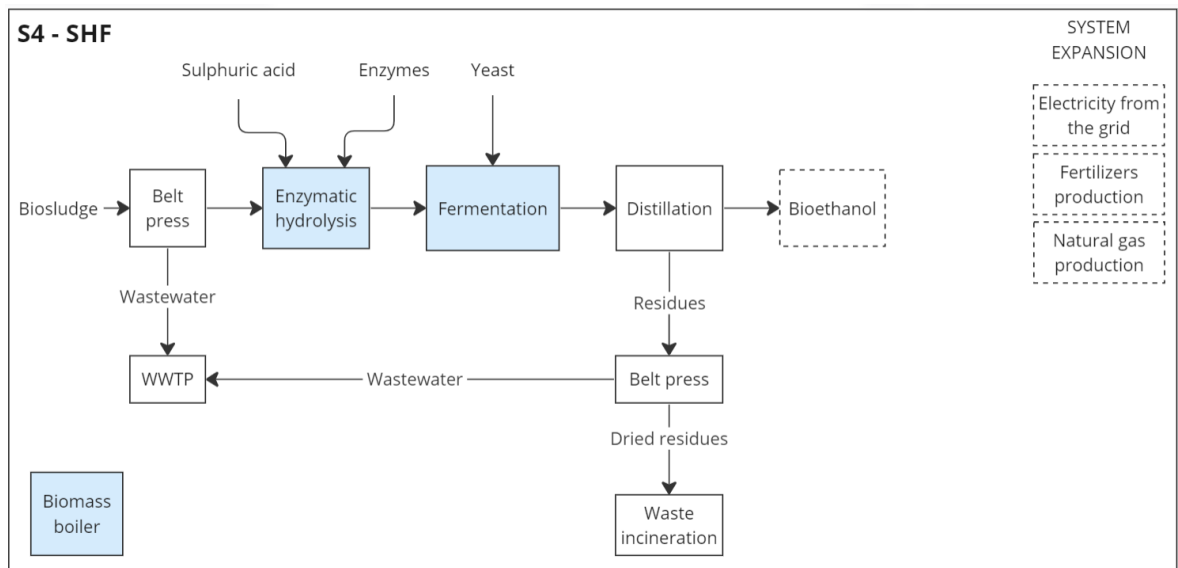
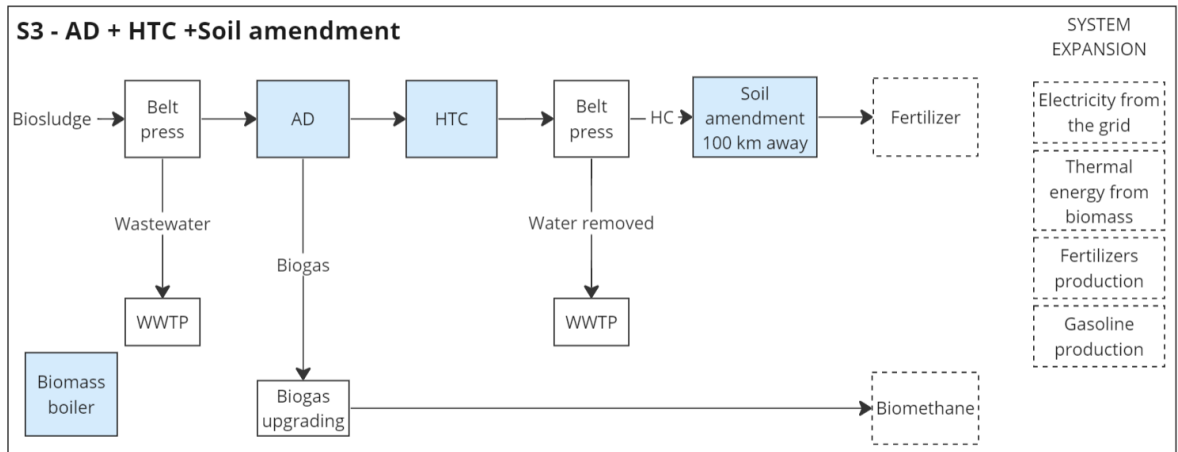


Figure 8. (continued)

4.2. Inventory analysis

Data inventory was built using different types of sources. Specific data on biosludge incineration and HTC were collected from a previous study that analyses the potential energy efficiency from hydrothermal carbonization and incineration of biosludge in Nordic pulp and paper mills (Saari et al., 2022). An on-going study on P&P biosludge anaerobic digestion provided experimental data on biosludge characteristics, AD yield, digestate HTC, and products composition. Data for landfilling of ashes and residues, electricity mix in Finland and wastewater treatment were sourced from Sphera database, as well as production of most of the chemicals. Finland was chosen as a location when data were available, otherwise Europe was chosen. Other additional data were retrieved from literature. Appendices 1 to 6 present the life cycle inventory of each scenario.

4.2.1. General assumptions

Based on (Saari et al., 2022), it is assumed that before each treatment method, biosludge is mechanically dewatered using a belt press. Even though mechanical dewatering could achieve dry content below 85%, biosludge in operating mills is only dewatered to around 8% dry solids to ensure an easy handling in pipes. Moisture content of biosludge is thus assumed to be decreased from 97% to 92%. The electricity required for dewatering is assumed to be 1 kWh per ton of water removed (Heimersson et al., 2014; Stoica et al., 2009). According to (Mohammadi et al., 2019a), 9 kg of polymer is also needed to dewater 1 ton of dry biosludge. Environmental impacts of polyacrylamide polymer production are sourced from (Braun et al., 2022).

Flue gas treatment and wastewater treatment are used on site to reduce the emissions of each conversion technology into the atmosphere and the hydrosphere. The environmental impacts related to the construction and maintenance of the infrastructures were not included in the study, based on the assumption that these are amortized over a reasonably long lifetime (Mohammadi et al., 2019a, 2019b). The impacts of transport from one treatment area to another within the mill site are not included either. Carbon sequestration is not considered in this study to avoid double counting with system expansion and biogenic emissions.

4.2.2. System 0 - Incineration

The baseline system of this study is based on (Saari et al., 2022), and more particularly on “NorInt” mill. In P&P mills, there are two alternatives for sludge incineration. Sludge can be incinerated with black liquor in a recovery boiler, or it can be incinerated with solid biomass residues (around 88% bark and 12% softwood fines) in a biofuel boiler (Stoica et al., 2009). The second option is less energy efficient but is often preferred as it prevents the system from introducing the non-process elements present in biosludge to the chemical recovery cycle (Saari et al., 2022). At “NorInt” mill, biosludge is dewatered and then sent to a circulating fluidized bed biomass boiler to be combusted with other waste from the mill, 42,768 kg of bark and 4,271 kg of fines for each tonne dry of biosludge. Characteristics of different feedstocks are given in **Table 5**. Around 90% of the heat available from the fuel is converted to steam. The steam generated by the boiler is used to generate electricity and heat for the mill consumption, steam turbine efficiency is estimated to be 33% (Breeze, 2014). The excess energy generated is then sold to replace electricity from the grid and heat produced from biomass combustion. Biosludge, bark and fines are the only fuels required during the operation of the boiler (Saari et al., 2022). Additional fuels are required for starting-up the boiler, but this is not included in this study. Input, outputs and emissions resulting from the combustion of biosludge, hydrochar, bark and fines in each scenario are given in Appendix 6. Steam flow, feedwater flow, flue gas mass flow and boiler capacity were calculated internally based on fuel mass flow, composition, and heating value. Operating data of the boiler were taken from (Saari et al., 2022), with an excess air ratio of 1.20.

Table 5. Characteristics of feedstocks for combustion (Lab tests; Martinez et al., 2021; Raatikainen, 2020; Zhang and Wang, 2013; Mannarino et al., 2022; Banks 2020).

Parameter	Unit	Biosludge	Bark	Fines	Hydrochar
Moisture	%	92.00	51.50	35.00	6.39
Ash	% dry	21.26	1.70	1.20	17.86
Volatile content	% dry	65.22	71.50	71.50	59.70
C	% dry	41.86	53.45	52.19	46.01
H	% dry	5.55	5.80	6.31	5.82
O	% dry	24.22	37.51	39.74	23.95
S	% dry	2.50	0.11	0.01	2.25
N	% dry	4.61	0.86	0.55	4.13
LHV _{db}	MJ/kg DS	18.50	19.60	19.70	23.10
Mass yield	% dry	-	-	-	82

Emissions resulting from combustion process were either calculated internally or extracted from the literature. Flue gas analysis was estimated using feedstocks characteristics (**Table 5**). The major pollutants that were considered in this study are (1) particulate matter, (2) metals, (3) carbon dioxide, (4) nitrogen oxides, and (5) sulphur dioxide (EPA, 1995). Limit values for dry flue gas emissions in Finland are given by (ECLI:FI:KHO:2017:T3829): sulphur dioxide 200 mg/m³, nitrogen oxides (NO₂) 150 mg/m³, particulate matter 20 mg/m³, carbon monoxide 250 mg/m³. Air pollution control technologies are sometimes needed to meet those limit values.

- (1) Wet scrubber is used for particulate matter control. It requires several chemical for pH adjustment and operation: sodium hydroxide (NaOH), sulphuric acid (H₂SO₄), sodium chloride (NaCl), hydrochloric acid (HCl), calcium chloride (CaCl₂) and sodium persulfate (Na₂S₂O₈) (Larsen et al., 2010; Mohammadi et al., 2019b).
- (2) Metal emissions are dependent on metal content in the feedstock, fuel bed temperature and level of particulate matter control (United States Environmental Protection Agency, 1995). Transfer coefficients of metals from sludge to exhaust gas are given by (Heimersson et al., 2014). Heavy metals concentration in biosludge and hydrochar are respectively extracted from (Mohammadi et al., 2019b) and (Eskandari et al., 2019). Metals emissions resulting from the combustion of bark and fines are not included in the model.
- (3) It is commonly assumed that 99% of the carbon in the fuel is converted to CO₂, the carbon not converted into CO₂ is going to bottom ash, the amount of CO produced is considered insignificant compared to the amount of CO₂ produced (United States Environmental Protection Agency, 1995). CO₂ emissions from combustion are biogenic.
- (4) The oxidation of nitrogen into NO_x occurs through a complicated oxidation-reduction reaction process (Chiang & Gao, 2022). According to (Mohammadi et al., 2019b), the combustion of one ton dry of biosludge emits 310g of NO₂ and 17.6g of NH₃, after denitrification. Selective non catalytic reduction is used for denitrification, the process requires 1.62 kg of ammonia. However, not only biosludge is combusted in each scenario, so it is assumed that those values are proportional to the amount of

nitrogen content in fuel. N₂O emissions calculations are based on emissions factors provided by (Guendehu et al., 2006): 450 g N₂O / t waste, on a wet basis.

- (5) Concerning SO₂ emissions, it is assumed that 2% of the sulphur content in fuel is released to the atmosphere as SO₂ after flue gas treatment (Havukainen et al., 2022). Less than 2% of SO₂ is usually converted to SO₃ (Chiang & Gao, 2022), so SO₃ emissions are neglected in this study. Sodium hydroxide (NaOH) is used as a neutralisation agent for SO₂ in alkaline scrubbing (Mohammadi et al., 2019b).

Residual bottom ash from combustion process is assumed to be 17% of the ash content in the fuel (Mohammadi et al., 2019a). Bottom ash is landfilled, which is a common practice in Nordic countries (Mohammadi et al., 2019b). All heavy metals that were not transferred to exhaust gas end up in the ash.

4.2.3. System 1 - HTC + incineration

In this scenario, biosludge is converted in an HTC plant into hydrochar, which is then combusted in the boiler for energy recovery. Hydrochar yield varies from 0.75 to 0.93, with an average value of 0.82, which was chosen for this study (Banks, 2021; Mannarino et al., 2022; Saari et al., 2022). Hydrochar characteristics are given in **Table 5**.

Hydrothermal carbonization of biosludge requires electric and thermal energy. It was found in literature that electricity demand for motor drives of pumps and screws, lighting, space heating can vary from 120 kWh/t_{dry} (Mohammadi et al., 2019a) to 283 kWh/t_{dry} (Raatikainen, 2020). The value of 212 kWh/t_{dry} was thus chosen. During the starting phase, an electric heater is also used temporarily. (Lucian & Fiori, 2017) researched hydrothermal carbonization energy consumption for different feedstocks and reaction conditions. They reported an average thermal energy consumption of 310 kWh/kg_{feedstock}, which accounts for 3,875 kWh/t_{dry} of biosludge. (Mohammadi et al., 2019a) rather suggests a thermal energy demand of 1,200 kWh/t_{dry} of biosludge. The average value of 2,538 kWh was chosen for this study.

The emissions resulting from hydrothermal carbonization of biomass, and especially biosludge from P&P industry, are not well reported in literature. In an environmental life cycle assessment of HTC of sewage sludge, (Mannarino et al., 2022) estimated the CO₂

biogenic emissions of the HTC process to be 4.57×10^{-2} kg/t_{feedstock} and CO emissions of 2.53×10^{-3} kg/t_{feedstock}. (Mohammadi et al., 2019a) did not consider any CO and CO₂ emissions, but only CH₄ emissions of 156 g/t_{dry} of biosludge. When analysing the emissions related to the HTC process of poultry litter, (Mau & Gross, 2018) confirmed the low emissions of CO₂, emissions of CH₄, and absence of emissions of NH₃ and N₂O, compared to biochar production through pyrolysis.

After HTC treatment, the slurry obtained is dewatered using a belt press. Electricity consumption is assumed to be 20 kWh_{el}/t_{dry} (Saari et al., 2022). Hydrochar combustion takes place in the same boiler where biosludge would otherwise be combusted, the same amounts of bark and fines as in S0 are combusted as well. Appendix 6 details mass flows associated with the incineration of hydrochar, bark and fines in scenario 1.

The treatment of HTC filtrate in a wastewater treatment plant enables to supply part of the nutrient required for P&P wastewater treatment (Hämäläinen et al., 2022), but this impact was not including in the study.

4.2.4. System 2 - HTC + soil amendment

In system 2, biosludge is also converted to hydrochar through hydrothermal carbonization. However, instead of being incinerated in the boiler, hydrochar is used as a soil amendment in forest. The location of application is assumed to be 100km away from the mill. The emissions to soil that results from hydrochar application on soil (Cadmium, chromium, copper, nickel, lead and zinc) are based on (Mohammadi et al., 2019a). This process makes it possible to avoid the use of conventional fertilizers, as reported in (Mohammadi et al., 2019a). In system 2, 3 and 4, only bark and fines are being incinerated in the biomass boiler. Mass flows of this process are detailed in Appendix 6.

4.2.5. System 3 - AD + HTC + Soil amendment

In system 3, biosludge is collected from the wastewater treatment plant, dewatered with a belt filter and then treated in an anaerobic digester to generate biogas. Biogas is a mixture of

methane, carbon dioxide, and impurities. An upgrading unit is thus necessary to produce biomethane, which can be used as vehicle fuel to replace natural gas (Heimersson et al., 2017). A common method for biogas upgrading is water scrubbing. The principle is based on CO₂ absorption into water under high pressure conditions. The CO₂ is then released back into the atmosphere (Mohammadi et al., 2019b). Those CO₂ emissions are assumed to be biogenic. The amount produced is calculated from the percentage of methane in the biogas (Møller et al., 2009), amounting for 64% (Mannarino et al., 2022). Biogenic CO₂ emissions are thus 36% of the methane production, on a volume basis. At standard temperatures and pressure, carbon dioxide weighs 1,98 kg/m³, so the anaerobic digestion of one tonne dry of biosludge produces 113 kg of biogenic CO₂.

The whole process (AD and biogas upgrading) requires 11.8 kWh of electricity (Mainardis et al., 2021; Mohammadi et al., 2019b), and 370 kWh of heat (Heimersson et al., 2017) per ton dry of biosludge treated. Heat demand is assumed to be met by the steam produced by the boiler. The products are biomethane and digestate. Biomethane potential is assumed to be 0.15 m³ CH₄/kg VS, specific methane potential is 90% of BMP and lower heating value of CH₄ is estimated at 10.23 kWh/m³, the final biomethane yield is thus 1,036 kWh/t dry of biosludge (Mainardis et al., 2021; Mannarino et al., 2022; Mohammadi et al., 2019b, Kinnunen et al, 2015). Digestate is produced at a rate of 0.57 t dry for 1 t dry of biosludge (Heimersson et al., 2017). Emissions generated from the process are methane due to leakage and nitrous oxide (Heimersson et al., 2017).

After anaerobic digestion, digestate is treated with HTC to produce hydrochar for nutrient recovery. It is assumed that the hydrochar yield from digestate is similar to undigested biosludge, carbon content is also assumed to be the same (Mohammadi et al., 2019b). Digestate treatment is thus similar to system 2, with a mass ratio of input feedstock into HTC of 0.57.

4.2.6. System 4 - Fermentation

In this scenario, biosludge is converted to bioethanol through separate hydrolysis and fermentation process. Data for system 4 were retrieved from literature. (Chen et al., 2014) simulated the conversion of P&P sludge to ethanol to analyse the economic feasibility of the

process. In this study, after being dewatered, biosludge is going to enzymatic hydrolysis. This step requires enzymes and sulphuric acid, respectively 921 kg and 716 kg for 1 ton of dry sludge (Chen et al., 2014). As it is very common for cellulosic ethanol production, cellulase is used in this scenario. The environmental impacts related to cellulase production are taken from (Gilpin & Andrae, 2017). After enzymatic hydrolysis, hydrolysed biosludge is going to fermentation and converted to bioethanol using yeast. 2.5 kg of yeast are needed for the conversion of one ton of substrate (Dunn et al., 2012). Thermal and electric energy required for the production of yeast are retrieved from (Dunn et al., 2012). The greenhouse gas emissions resulting from the production of 1 kg of yeast are 3.2 kg CO₂ eq. (Soam et al., 2016). Once fermentation is done, the fermented biosludge is going to distillation to extract ethanol. The resulting residues are dewatered with a belt press and sent to incineration. The water removed is mainly recycled into the process, the rest of it goes to the wastewater treatment plant on site (Chen et al., 2014). It is assumed that bioethanol is replacing gasoline as a transport fuel.

4.2.7. System expansion

To assess the environmental benefits and costs of each co-product generated in the different scenarios, system expansion was used in this study. This approach is a commonly used method to analyse the end-of-life of a system (Mohammadi et al., 2019b). Two methods can be chosen for system expansion to ensure the comparability of the different scenarios analysed. First, the avoided emissions from the generation of co-products to replace conventional products can be accounted as credits, it is called system expansion with displacement (U.S. DOE, 2022). It is a common practice in LCA studies of waste management systems (Havukainen et al., 2022; Mohammadi et al., 2019b, 2019a; Sebastião et al., 2016). System expansion can otherwise be modelled by including in the baseline scenario the additional processes necessary to ensure an equivalent function with the studied scenario. In studies with multiple scenarios, all systems must provide an equivalent function. The first approach can sometimes result in net negative total emissions, which does not imply an uptake of emissions but rather a better environmental performance compared to the baseline scenario (U.S. DOE, 2022). In the present study, the second approach was chosen to account for the net positive environmental impact of biosludge treatment.

Five resources are produced in this study: (1) electricity and (2) heat from the boiler and fermentation residues incineration, (3) fertilizer from the use of hydrochar as soil amendment, (4) biomethane from anaerobic digestion and (5) bioethanol from fermentation process. Each resource is respectively replaced by (1) electricity from the grid, (2) heat from biomass combustion, (3) N, P, K, Mg, Ca fertilizers, (4) natural gas and (5) gasoline. When a co-product is not produced in one of the scenarios, its replacement resource is used instead. **Table 6** details the resources produced and needed in each scenario.

Table 6. Inventory related to system expansion.

Resources produced at the mill	Unit	S0	S1	S2	S3	S4
Electricity*	kWh	34,671	36,345	35,698	35,740	35,932
Heat*	kWh	59,922	60,660	59,541	60,262	62,116
Fertilizer (hydrochar)	kg	0	0	871	566	0
Biomethane	kWh	0	0	0	1,036	0
Bioethanol	kg	0	0	0	0	173
Additional resources needed	Unit	S0	S1	S2	S3	S4
Electricity from the grid	kWh	1,675	0	648	605	413
Thermal energy from biomass	kWh	2,194	1,456	2,575	1,853	0
N fertilizer (NH ₄ NO ₃)	kg	6.53	6.53	0	2.28	6.53
P fertilizer (P ₂ O ₅)	kg	0.87	0.87	0	0.30	0.87
K fertilizer (K ₂ O)	kg	0.80	0.80	0	0.28	0.80
Mg fertilizer (MgO)	kg	0.13	0.13	0	0.05	0.13
Ca fertilizer (CaCO ₃)	kg	0.13	0.13	0	0.05	0.13
Natural gas	kWh	1,036	1,036	1,036	0	1,036
Gasoline	kg	109	109	109	109	0

The scenario in which the most electricity is generated is scenario 2, when hydrochar is incinerated instead of biosludge. In this system, the mill is able to sell 36,345 kWh of electricity. When less electricity is generated in the other scenarios, electricity is needed from the grid to reach the same amount of total electricity. Concerning fertilizers, it is assumed that hydrochar use as a soil amendment makes it possible to avoid the use of N fertilizer (NH₄NO₃), P fertilizer (P₂O₅), K fertilizer (K₂O), Mg fertilizer (MgO), Ca fertilizer (CaCO₃). Data about fertilizers replacement are retrieved from (Mohammadi et al., 2019a).

It is assumed that biomethane can replace the use of natural gas consumption mix in Finland. Biomethane production is expressed in energy unit to take into account the difference in calorific value compared to natural gas. Bioethanol can replace gasoline consumption mix at filling station in Europe. Ethanol lower heating value is estimated to be 26.8 MJ/kg, while gasoline heating value is estimated to be 42.5 MJ/kg (Malça & Freire, 2006). The substitution factor is therefore 0.63 kg of gasoline for 1kg of bioethanol.

4.3. Life cycle impact assessment

According to the ISO 14040 guidelines, life cycle impact assessment (LCIA) is comprised of the following steps: 1) selection of impact categories; 2) assignment of LCI results (classification); 3) calculation of category indicator results (characterisation), 4) normalization; and 5) weighting. While the first three steps are mandatory, the last two are optional and were not carried out in this study. LCIA was performed using LCA for Experts 10.7.0.183 software (GaBi) and Professional Database 2023 from Sphera (Sphera, 2023). The mid-point impact method Recipe 2016 was used as the selected LCIA method and the hierarchist perspective (H) was chosen to evaluate environmental impacts over a time horizon of 100 years. Replacing biosludge incineration by other thermochemical and biological treatments aims at increasing energy and nutrient recovery by producing hydrochar, biofuels and fertilizer, reducing environmental and health impacts resulting from pollutants emissions from combustion process, and immobilizing heavy metals (Mohammadi et al., 2019b, 2019a). Therefore, the selected impact categories include climate change (including biogenic carbon) (CC), fine particulate matter formation (FPMF), terrestrial acidification (TA), freshwater eutrophication (FE), terrestrial eco-toxicity (TET), human toxicity (cancer) (HTc), marine eutrophication (ME). The impact categories are described in Table 7. A contribution analysis was carried out to assess the contribution of each sub-systems to the different environmental impact categories.

As mentioned in section 3.2, biogenic emissions are not always included in environmental impact studies. Yet, (Cherubini et al., 2012) analysed the contribution of biogenic GHG to global warming potential and concluded that in the case of forest wood harvested in areas affected by seasonal snow cover, biogenic GHG emissions are both contributing to

global warming because of their longer residence time in the atmosphere compared to fast growing species and contributing to cooling because of the increased snow surface after harvest. Given the mixed impact of biogenic carbon emissions in the case of this study, an equivalency factor of 1 is attributed to them for the measure of climate change impact (1 kg biogenic CO₂ = 1 kg CO₂ eq.) and their contribution will be further analysed in the results. Both biogenic carbon dioxide and methane are included in the study.

Table 7. Selected impact categories of the ReCiPe 2016 methodology (Havukainen et al., 2022).

Impact category	Unit	Description
Climate change	kg CO ₂ eq. to air	Impact of greenhouse gas emissions on the global warming
Fine particulate matter formation	kg PM _{2.5} eq. to air	Air pollution which leads to aerosol formation in the atmosphere
Terrestrial acidification	kg SO ₂ eq. to air	Deposition of inorganic substances causing change in acidity in the soil
Freshwater eutrophication	kg P eq. to freshwater	Discharge of nutrients to freshwater causing rise in nutrient level
Terrestrial eco-toxicity	kg 1,4-DCB eq. to industrial soil	Damage caused to ecosystems by a chemical emission
Human toxicity: cancer	kg 1,4-DCB to urban air	Damage caused to human health by a chemical emission
Marine eutrophication	kg N eq. to marine water	Discharge of nutrients to sea water causing rise in nutrient level

4.4. Uncertainty analysis

LCA studies are subjects to three main types of uncertainty (Clavreul et al., 2012). Parameter uncertainties are due to the data variability in space and time, erroneous measurements, wrong estimations. Scenario uncertainties are due to the choices related to scenarios. Model uncertainties arise from the mathematical equations used to model reality. Table 8 gives a non-exhaustive list of the sources of uncertainties in this study. In LCA of waste management systems, the amount and preciseness of data are still limited, thus LCA-modelling requires many assumptions that leads to different types of uncertainties (Clavreul et al., 2012).

Table 8. Sources of uncertainties in this study (Adapted from Clavreul et al., 2012).

Process	Model uncertainty	Scenario uncertainty	Parameter uncertainty
General	Linearity of emissions	System boundaries (exclusion of on-site transportation and capital goods) Database for energy, landfilling and material productions	
Impact assessment	Recipe 2016 model to calculate characterisation factors Linearity of response	Time horizon of impact characterisation (100 years)	Characterisation factors
Biosludge, hydrochar, bark, fines, digestate composition	Models for some substances' fate during HTC and AD		Chemical composition and heating value
Transport		Choice of database for truck and fuel	Distance
Incineration	Models for flue gas emissions Model for heavy metals' fate	Choice of boiler and flue gas control technologies	Electricity and heat recovery, consumption of chemicals, water, and sand Output of pollutants and ashes
Alternative treatment methods	Model for residues incineration	Choice of technology	Yields Emissions due to each treatment Enzymes and yeast consumption and emissions
System expansion	Model for fertilizer substitution	Choice of products utilisation Choice of substitution	

The robustness of the results was assessed in two parts. First, an uncertainty propagation was performed to calculate the uncertainty of the results considering all input uncertainties. The Monte Carlo analysis was chosen because it is the most common method used and results are calculated in a reasonable amount of time (Clavreul et al., 2012). Standard deviations were directly obtained from literature, calculated based on multiple values found in literature or estimated based on Ecoinvent 3.0 recommendations (Weidema et al., 2013). Second, sensitive inputs were identified using a sensitivity analysis. Some inputs were selected and modified one at a time to evaluate their influence on the results. This type of sensitivity analysis is called perturbation analysis. Sensitivity ratio (SR) can then be calculated

according to (Clavreul et al., 2012). It is the ratio between the relative change of the total result and the relative change of the individual parameter. **Table 9** details the baseline value of the parameters chosen for sensitivity analysis, their range and incertitude.

Table 9. Key parameters chosen for the sensitivity analysis.

Parameter	Unit	Baseline	Range	SD (%)	Δp
Hydrochar yield	kg dry HC/kg dry BS	0.82	0.75 - 0.93	12 %	14 %
Biomethane yield	kWh/t dry BS	1036	444 - 1269	34 %	23 %
Bioethanol yield	kg/t dry BS	173	149 - 197	15 %	14 %
Cellulase dosage	kg/t dry BS	33	9 - 100	135 %	202 %
CC impact of cellulase	kg CO ₂ eq / kg CE	15.0	2.3 - 25.4	93 %	69 %
TA of cellulase	g SO ₂ eq / kg CE	171	26 - 415	163 %	143 %
FE of cellulase	g P eq / kg CE	5.6	2.2 - 11.1	84 %	98 %
WW from HTC	kg/t dry BS	12,460	11,214 – 13,706	10%	10%
WW from initial dewatering	kg/t dry BS	20,833	18,750 – 22,916	10%	10%
C% in boiler feedstock S0	kg/FU	12,376	11,138 – 13,614	10%	10%
C% in boiler feedstock S1	kg/FU	12,098	10,888 – 13,308	10%	10%
C% in boiler feedstock S2,3,4	kg/FU	11,957	10,761 – 13,153	10%	10%

CC: climate change; CE: cellulase enzyme; FE: freshwater eutrophication; HTC: hydrothermal carbonization; SD: standard deviation; TA: terrestrial acidification; WW: wastewater; Δp : relative change of the parameter (maximum absolute value).

5. Results

Table 10 and **Figure 9** present the impact assessment results for CC, FPMF, TA, FE, TE, HTc, ME of the studied scenarios, as well as the relative changes compared to the baseline scenario S0. All scenarios show an overall decrease in climate change impact. With a maximum decrease of 3.37 tonnes CO₂ eq. in S2 compared to S0. The changes in other impact categories are more diverse. Scenarios 2, 3 and 4 show a reduction in fine particulate matter formation, terrestrial acidification and terrestrial ecotoxicity, while freshwater eutrophication, human toxicity and marine eutrophication are increased compared to the baseline scenario. Out of those three scenarios, S3 shows the best performance in four out of seven categories. As for scenario 4, despite showing the best environmental performance in TE, HTc and ME categories, fine particulate matter formation, terrestrial acidification and freshwater eutrophication are greatly increased compared to S0. Freshwater eutrophication in S4 is nine times higher than in the baseline scenario, with a value of 207 g P eq. compared

to 23 g P eq. in S0. Freshwater eutrophication is increased in all alternative scenarios, compared to the baseline scenario. **Table 10** also present the standard deviations of all scenarios. Standard deviations were calculated with a Monte Carlo analysis on LCA for Expert software. FPMF, TA and TE impact categories are subject to high uncertainties (> 45%), as well as freshwater eutrophication in scenario 4 (104%).

Table 10. LCIA results.

Results		S0	S1	S2	S3	S4
Climate change, incl. biogenic carbon (CC)	kg CO2 eq.	53,516	50,584	50,145	50,232	50,944
Fine Particulate Matter Formation (FPMF)	kg PM2.5 eq.	0.79	0.51	0.72	0.62	2.36
Terrestrial Acidification (TA)	kg SO2 eq.	2.49	1.64	2.27	1.96	8.05
Freshwater Eutrophication (FE)	kg P eq.	0.023	0.031	0.031	0.028	0.207
Terrestrial ecotoxicity (TE)	kg 1,4-DB eq.	395	341	245	190	167
Human toxicity, cancer (HTc)	kg 1,4-DB eq.	2.26	2.67	2.91	2.54	2.00
Marine Eutrophication (ME)	kg N eq.	0.070	0.081	0.082	0.075	0.056
Relative changes compared to S0		S0	S1	S2	S3	S4
Climate change, incl. biogenic carbon	kg CO2 eq.	0	-5 %	-6 %	-6 %	-5 %
Fine Particulate Matter Formation	kg PM2.5 eq.	0	-35 %	-8 %	-22 %	199 %
Terrestrial Acidification	kg SO2 eq.	0	-34 %	-9 %	-21 %	223 %
Freshwater Eutrophication	kg P eq.	0	33 %	33 %	18 %	780 %
Terrestrial ecotoxicity	kg 1,4-DB eq.	0	-14 %	-38 %	-52 %	-58 %
Human toxicity, cancer	kg 1,4-DB eq.	0	19 %	29 %	13 %	-11 %
Marine Eutrophication	kg N eq.	0	16 %	17 %	8 %	-21 %
Standard deviations		S0	S1	S2	S3	S4
Climate change, incl. biogenic carbon	%	7 %	7 %	7 %	8 %	7 %
Fine Particulate Matter Formation	%	54 %	54 %	54 %	55 %	62 %
Terrestrial Acidification	%	54 %	54 %	54 %	55 %	63 %
Freshwater Eutrophication	%	7 %	8 %	8 %	7 %	104 %
Terrestrial ecotoxicity	%	45 %	45 %	45 %	46 %	46 %
Human toxicity, cancer	%	26 %	26 %	26 %	27 %	27 %
Marine Eutrophication	%	17 %	17 %	17 %	17 %	18 %

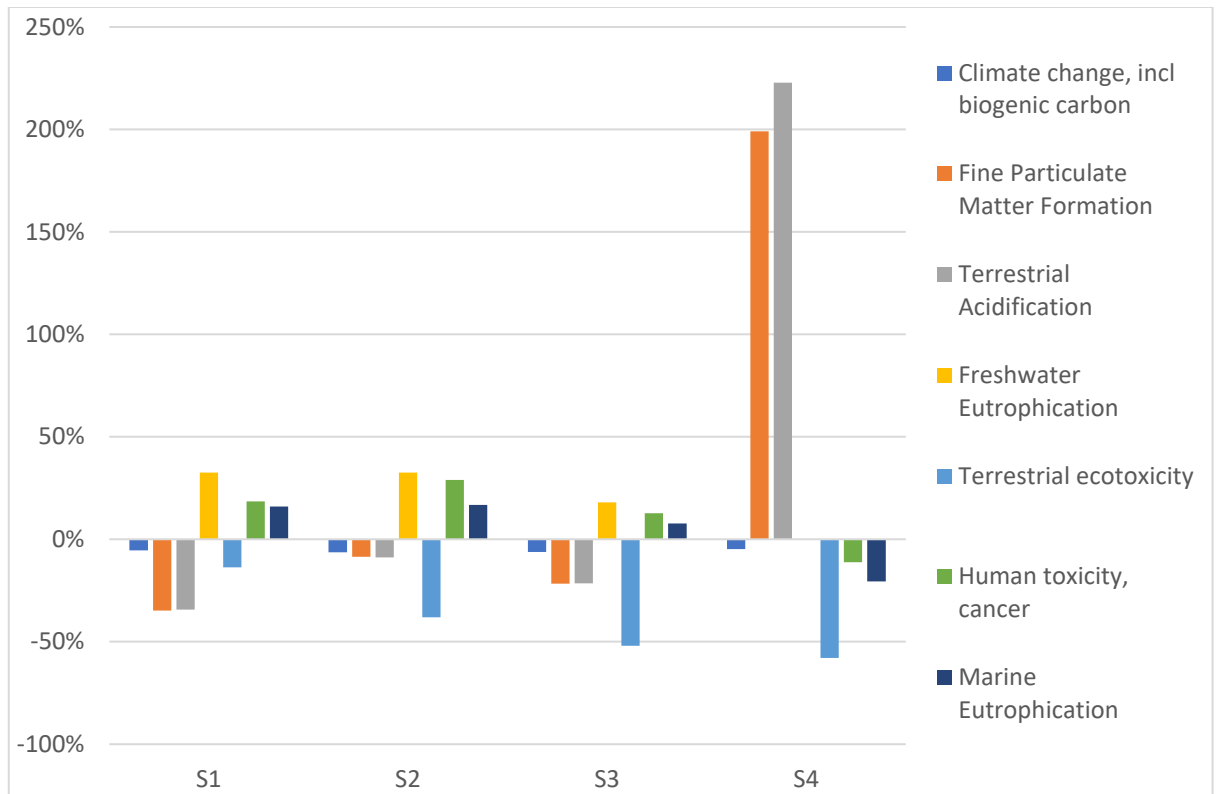


Figure 9. LCIA contribution assessment of each scenario compared to the baseline scenario (S0). Freshwater eutrophication relative change of S4 compared to S0 is not displayed for a better visibility, because this value is considerably higher than the other relative changes (780%).

5.1. Contribution analysis

Figure 10 shows the relative influence of each sub-system to the total environmental impacts. Standard deviations are displayed with error bars on the figure. Appendix 7 gives detailed results of the contribution analysis. Climate change impact of bark and fines combustion are not included on Figure 10.

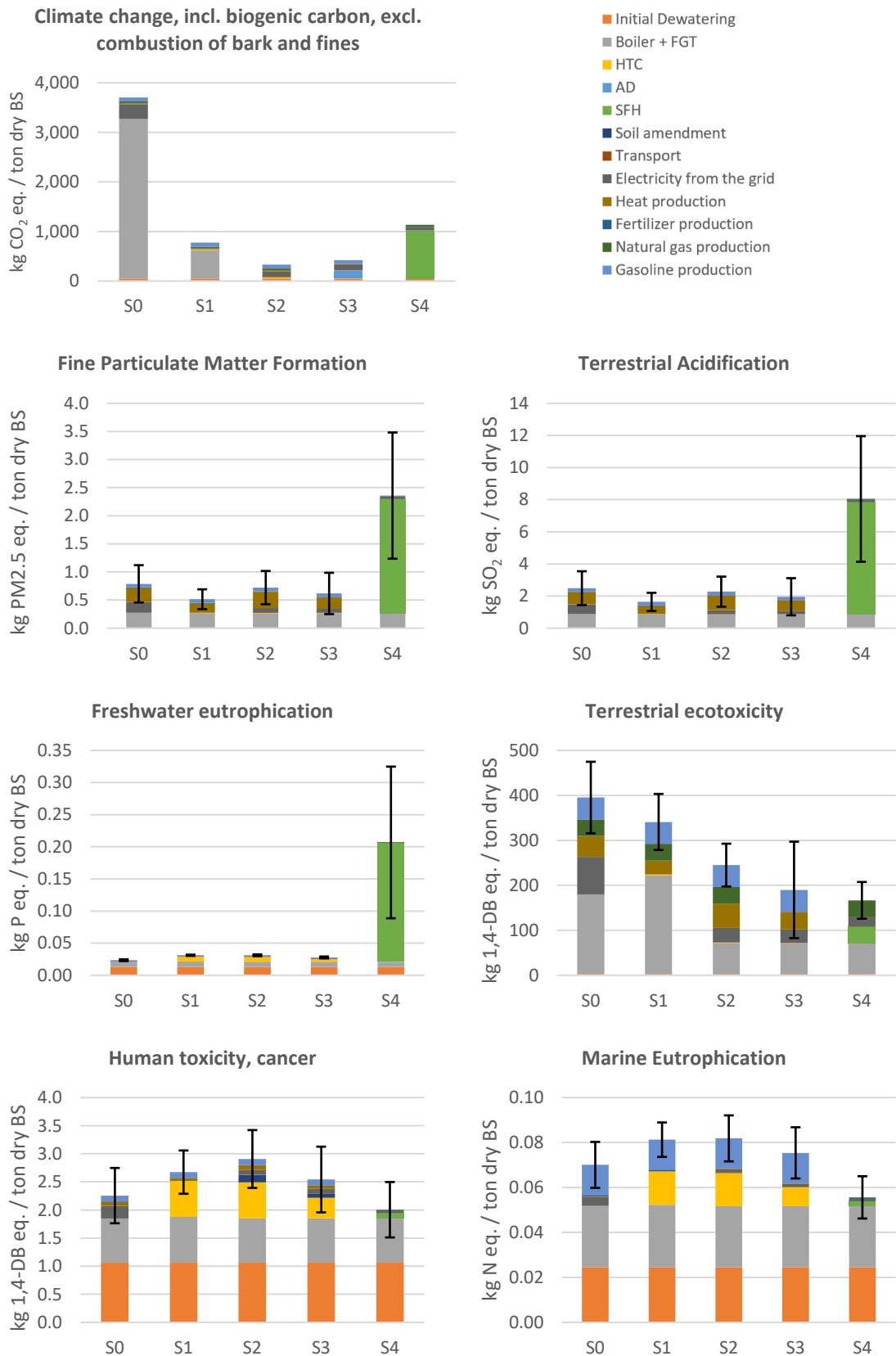


Figure 10. LCIA contribution assessment for the selected impact categories (standard deviation of the results are shown with error bars). AD: anaerobic digestion; BS: biosludge; FGT: flue gas treatment; HTC: hydrothermal carbonization; SFH: separate hydrolysis and fermentation.

Climate change impact category is greatly affected by the emissions resulting from the biomass boiler and treatment of flue gas. In each scenario, this sub-system accounts for more than 97% of the total climate change impact, most of it being caused by biogenic CO₂ emissions from flue gas (> 84%). To understand better the environmental benefits of alternative biosludge treatment, Figure 10 presents the climate change impact of each scenario, excluding the emissions related to the combustion of bark and fines (49,810 kg CO₂ eq.). The combustion of biosludge in S0 amounts to 3,220 kg CO₂ eq., thus accounting for 6.2% of the total emissions of the boiler and flue gas treatment, including bark and fines combustion. This result is in accordance with the ratio of dry solids input in the boiler, for which biosludge stands for 4.3%. When bark and fines incineration is excluded from the results, biosludge incineration contributes to 87% of the total climate change impact in S0. The high contribution of boiler and flue gas treatment to climate change is in line with (Mohammadi et al., 2019b). (Pfadt-Trilling et al., 2021) also evaluated the contribution of incineration to 90% of the emissions of waste-to-energy systems. In S1, the combustion of hydrochar instead of biosludge reduces the emissions of the boiler by 2,657 kg CO₂ eq. (-83%) and avoids the emissions of 289 kg CO₂ eq. due to the additional electricity generated. Separate hydrolysis and fermentation process in S4 contributes to 85% of climate change emissions, when bark and fines incineration is excluded.

The increased environmental impacts in freshwater eutrophication, human toxicity, and marine eutrophication for S1, S2 and S3 compared to S0 are due to the emissions related to the HTC process. A great amount of wastewater is generated during HTC (12.5 t for one ton dry of biosludge), the treatment of this water contributes to freshwater eutrophication, human toxicity (cancer), and marine eutrophication. HTC filtrate treatment in WWTP stands for more than 68% of the total emissions of the HTC process in all impact categories and reaches 99% of the total emissions of FE, HTc, and ME. The high contribution of wastewater treatment to human toxicity (cancer) in sludge treatment has also been demonstrated by (Mohammadi et al., 2019b). Not only HTC treatment is concerned by wastewater treatment impact but also the initial dewatering of biosludge (Figure 10).

Scenario 4 has been analysed in more detail to identify the sub-processes resulting in the high increase in fine particulate matter formation, terrestrial acidification and freshwater eutrophication. In those three impact categories, enzyme production for enzymatic hydrolysis of biosludge contributes respectively to 70%, 70% and 89% of the total emissions of the scenario. However, those results are subject to great uncertainty as shown on Figure 10 and Table 10. Even when considering the range of the results, S4 could not outperform the other scenarios in those three categories. Enzyme production contributes to 51% of the climate change impact of SHF process (excluding boiler and system expansion). This result is in line with (Soam et al., 2016) that assessed an enzyme production contribution of 54% to 57% to GHG emissions of ethanol production. (Sebastião et al., 2016) also pointed out the high contribution of enzyme production to the environmental impact of bioethanol production, with a contribution of over 80% to global warming potential. Considering a lower heating value of 26.8 MJ/kg for bioethanol (Malça & Freire, 2006), the GHG emissions associated with bioethanol production are estimated to be 220 g CO₂ eq./MJ. This result is above the typical values found in literature: (Sebastião et al., 2016) estimated a global warming potential of 73 g CO₂ eq./MJ. However, the scope of (Sebastião et al., 2016) does not include biogenic carbon emissions and end-of-life of fermentation residues. Yet, residues incineration accounts for 41% of GHG emissions of SHF process in the present study. According to (Soam et al., 2016), GHG emissions of cellulosic ethanol ranges from 8 g CO₂ eq./MJ to 76 g CO₂ eq./MJ (for various plant-based feedstocks). Those results usually allocate energy production from the process as negative emissions and do not include biogenic carbon emissions, which can explain the higher value for the present study. When energy production in SHF process of S4 is accounted as credits (-11 kg CO₂ eq. for heat generation and -26 kg CO₂ eq. for electricity generation), the emission factor of bioethanol production amounts to 212 g CO₂ eq./MJ.

5.2. Sensitivity analysis

The parameters chosen for sensitivity analysis can be categorized in two types: (1) yields of energy and nutrients carriers and (2) parameters identified as high contributors in the contribution analysis. Each parameter was analysed individually to assess the sensitivity of the results to its variation. The sensitivity ratios (SR) of each parameter are displayed on Table 11. According to (Havukainen et al., 2022), parameters with SR values below 0.2 have

a low influence on the results, while parameters with SR values above 0.8 have a great influence on the overall results.

Table 11. Sensitivity ratio for the selected parameters.

Parameter	CC	FPMF	TA	FE	TE	HTc	ME
S0 Incineration							
Hydrochar yield	0.01	0.10	0.11	0.00	0.09	0.03	0.03
Biomethane yield	0.00	0.04	0.04	0.00	0.24	0.02	0.01
Bioethanol yield	0.00	0.08	0.09	0.09	0.13	0.06	0.19
C% in boiler feedstock	0.82	0.00	0.00	0.00	0.00	0.00	0.00
WW from initial dewatering	0.00	0.01	0.00	0.55	0.03	0.49	0.36
S1 HTC + Energy recovery							
Hydrochar yield	0.00	0.25	0.26	0.02	0.38	0.03	0.01
Biomethane yield	0.00	0.06	0.05	0.01	0.27	0.00	0.01
Bioethanol yield	0.00	0.12	0.13	0.07	0.15	0.05	0.17
C% in boiler feedstock	0.86	0.00	0.00	0.00	0.00	0.00	0.00
WW from HTC	0.00	0.00	0.00	0.26	0.00	0.26	0.18
WW from initial dewatering	0.00	0.02	0.00	0.42	0.00	0.41	0.31
S2 HTC + Nutrient recovery							
Hydrochar yield	0.00	0.12	0.13	0.02	0.15	0.05	0.01
Biomethane yield	0.00	0.04	0.04	0.00	0.38	0.02	0.01
Bioethanol yield	0.00	0.09	0.09	0.07	0.20	0.05	0.17
C% in boiler feedstock	0.86	0.00	0.00	0.00	0.00	0.00	0.00
WW from HTC	0.00	0.01	0.00	0.26	0.00	0.24	0.18
WW from initial dewatering	0.00	0.01	0.00	0.42	0.00	0.38	0.31
S3 AD							
Hydrochar yield	0.01	0.14	0.11	0.00	0.19	0.06	0.02
Biomethane yield	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Bioethanol yield	0.00	0.10	0.11	0.08	0.26	0.06	0.18
C% in boiler feedstock	0.86	0.00	0.00	0.00	0.00	0.00	0.00
WW from HTC	0.00	0.00	0.00	0.18	0.00	0.16	0.11
WW from initial dewatering	0.00	0.02	0.00	0.47	0.00	0.43	0.33
S4 Fermentation							
Hydrochar yield	0.00	0.03	0.03	0.00	0.17	0.07	0.04
Biomethane yield	0.00	0.02	0.01	0.00	0.58	0.02	0.00
Bioethanol yield	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cellulase dosage	0.01	0.70	0.70	0.90	0.00	0.00	0.00
Environmental impacts of cellulase production	0.01	0.87	0.88	0.77	0.00	0.00	0.00
C% in boiler feedstock	0.86	0.00	0.00	0.00	0.00	0.00	0.00
WW from initial dewatering	0.00	0.00	0.01	0.10	0.06	0.55	0.45

0.2 < SR < 0.8; SR > 0.8

C%: content of carbon; CC: climate change; FE: freshwater eutrophication; FPMF: fine particulate matter formation; HTC: hydrothermal carbonization; HTc: human toxicity, cancer; ME: marine eutrophication; TA: terrestrial acidification; TE: terrestrial ecotoxicity; WW: wastewater.

In all scenarios, the content of carbon in boiler feedstock has a SR over 0.8 in climate change category. Carbon content is indeed directly proportional to the carbon dioxide of the boiler. In scenario 4, cellulase dosage and environmental impact of cellulase production have a major impact on fine particulate matter formation, terrestrial acidification, and freshwater eutrophication. More precise results would be obtained with a site-specific study on cellulase need and origin in the case of biosludge fermentation on a pulp and paper mill. Several studies have analysed the potential of cellulase recycling for biorefineries (Jampana et al., 2021). (Gomes et al., 2016) demonstrated an enzyme saving of 54-60% in the case of cellulase recycling for bioethanol production. Cellulase recycling can not only reduce the need for cellulase in hydrolysis process but also decrease the environmental impact of its production and reduce enzyme cost for a more attractive industrial application (Q. Wang et al., 2016).

6. Discussion

This study differs from previous studies on pulp and paper biosludge treatment (Mohammadi et al., 2019a, 2019b; Sebastião et al., 2016) on two main aspects: (1) biogenic emissions have been accounting for in the evaluation of global warming potential; (2) system expansion is not based on co-products displacement (avoided emissions) but rather multiple products substitution to ensure an equivalent function in all scenarios. Those choices will be discussed in this section, as well as other relevant observations.

In Recipe 2016 impact method, biogenic CO₂ emissions are given an equivalency factor (BCEF) of 1 for their contribution to climate change impact category. However, (Cherubini et al., 2012) warns LCA practitioners that approximating biogenic CO₂ by a factor 0 or 1 can lead to misleading results. The climate change equivalency factor of biogenic emissions should be site-specific, depending on the climate of the area of harvest, feedstock species, and time horizons. For feedstock sourced in Nordic forests, BCEF is suggested to be 0.2 (global warming potential over 100 years). The factor has to be multiplied by 34.5 for biogenic CH₄ emissions (Cherubini et al., 2012). As stated by (Dong et al., 2021), “global

warming potential” impact category from CML method and “climate change” impact category from Recipe 2016 method can be converted to the same indicator using a conversion factor, with a high correlation. Therefore, the equivalency factor provided by (Cherubini et al., 2012) can be directly applied to climate change impact category in Recipe 2016 method. Figure 11 reveals the influence of biogenic carbon equivalency factor on climate change results. In this study, where BCEF is chosen to be 1, climate change results have been overestimated by 221% (average relative change for all scenarios), in comparison with a BCEF of 0.2 as recommended by (Cherubini et al., 2012). If biogenic carbon emissions were to be excluded of the scope of the study, climate change results would be underestimated by 55% (average relative change for all scenarios), in comparison with a BCEF of 0.2. With a BCEF of 0.2, climate change impact is reduced by 12% in scenarios S1 to S3 and 10% in S4 compared to the baseline scenario S0.

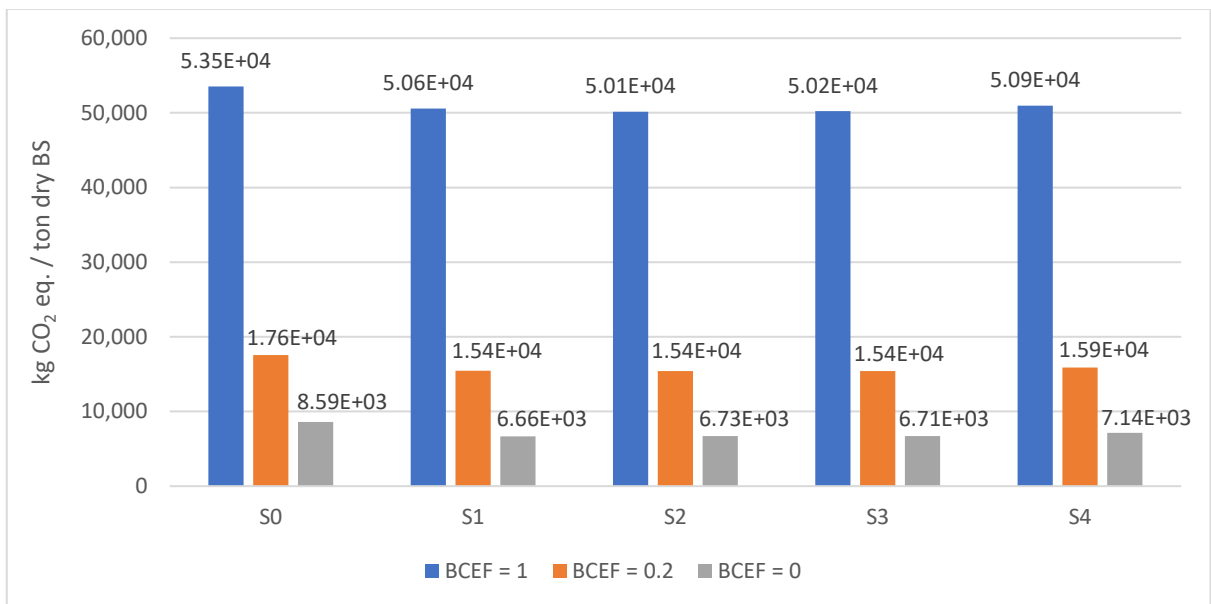


Figure 11. Influence of biogenic carbon equivalence factor on climate change [kg CO₂ eq.]. BCEF: Equivalency factor for biogenic carbon emissions.

System expansion modelling is a critical aspect of waste management LCA studies, the choice of modelling can greatly influence the results (Viau et al., 2020). The modelling of system expansion in this study makes comparison with literature results more complex because it differs from the most common practice that is substitution. However, (Tsalidis & Korevaar, 2020) observed a paradox related to substitution method with displacement as a

way of handling multifunctional processes in LCA studies. When a product-related functional unit is chosen and substitution method is applied, avoided emissions will be higher the lower the conversion efficiency of the technology. Therefore, low energy-efficient technologies can environmentally outperform more efficient technologies. The authors also warn LCA practitioners about the overestimation of climate change benefits due to energy credits. Accounting avoided emissions as credits can lead to negative overall emissions, which has to be considered carefully by policymakers as it does not account for emissions uptake (Pfadt-Trilling et al., 2021). In this study, the main objective of each system is to treat a certain amount of biosludge. The generation of co-products is a secondary objective, contributing to the optimisation of biosludge treatment. Therefore, choosing a fix mass of biosludge as the main functional unit, and the generation of co-products as a secondary functional unit, ensure the equity of each scenario.

To be able to compare the results of this study with other studies, Figure 12 presents climate change results excluding bark and fines treatment, biogenic emissions, and considering a system expansion with co-products displacement. Compared to the baseline scenario, scenarios S1 to S4 decrease the emissions by 111%, 107% 108% and 83% respectively. When considering the results on Figure 12, all scenarios reduce the climate change impact of biosludge handling. S1, S2 and S3 even demonstrate negative impact thanks to high emissions abatement. Those results are in accordance with (Mohammadi et al., 2019a) which estimated climate change impact of S3 around -150 kg CO₂ eq. For the comparison between S0 and S3, results differ from (Mohammadi et al., 2019b) because nitrous oxide emissions were included in the impact of biosludge incineration in this study, accounting for 1,675 out of 1,740 kg CO₂ eq. (Mohammadi et al., 2019b) estimated a climate change reduction of 391% between biosludge incineration and combined AD and pyrolysis. This result is similar to S3 results when excluding nitrous oxides emissions.

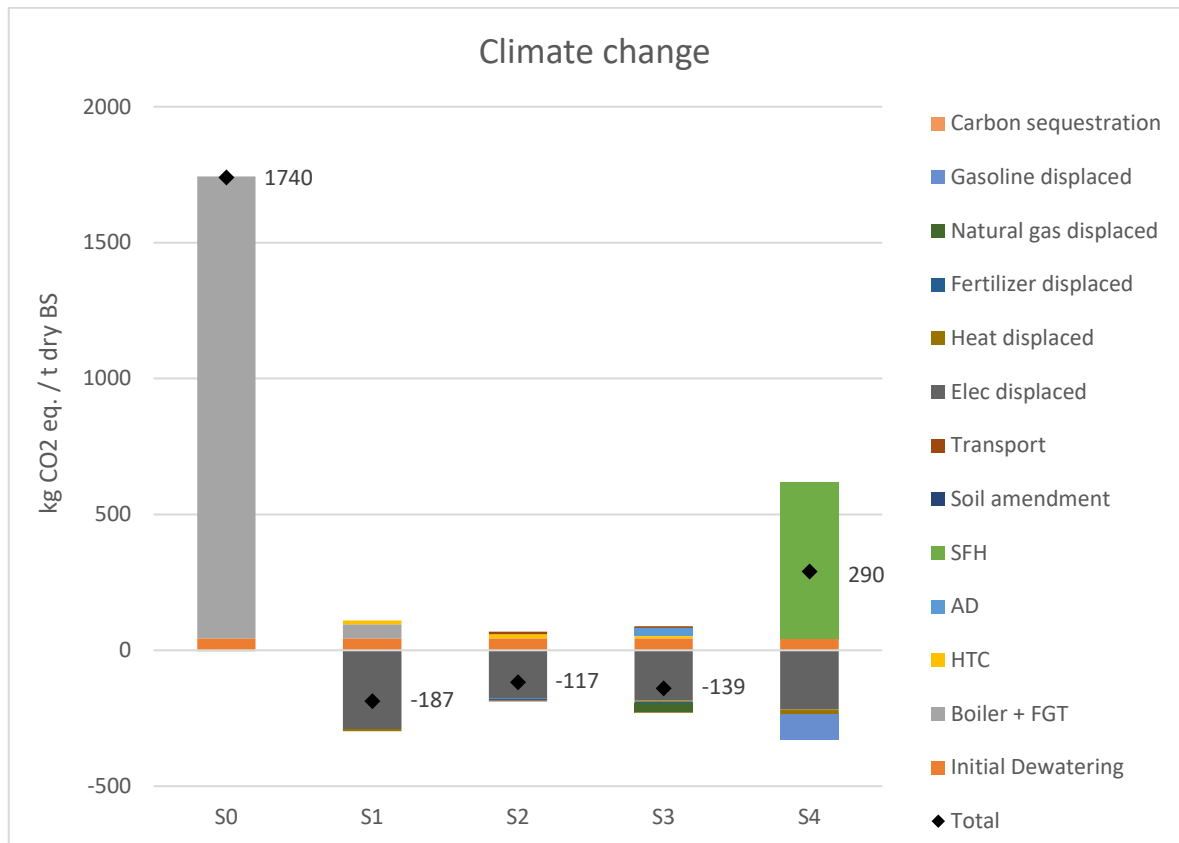


Figure 12. Climate change contribution assessment excluding bark and fines treatment, biogenic carbon emissions and modelling system expansion with co-products displacement. AD: anaerobic digestion; BS: biosludge; FGT: flue gas treatment; HTC: hydrothermal carbonization; SFH: separate hydrolysis and fermentation.

HTC filtrate treatment contributes greatly to the freshwater eutrophication, human toxicity and marine eutrophication of scenarios S1, S2 and S3 (> 99% of HTC process impact). To reduce the environmental impact of HTC filtrate, several studies have analysed the feasibility and advantages of alternative treatment methods: recycling the filtrate back into the HTC reactor (Banks, 2021; Mäkelä et al., 2018; Tasca et al., 2019; L. Wang et al., 2019) or producing biomethane through AD (Hämäläinen et al., 2021, 2022). Filtrate recirculation into the HTC reactor improves the mass and energy yields, as well as dewaterability, of hydrochar compared to HTC process without recirculation. Volatile matter of hydrochar is also decreased, enhancing carbon stability in soil. HTC filtrate has a higher COD concentration and biomethane potential than P&P mill sludge, respectively 8-12 fold higher and 5-6 fold higher than for a mixture of primary and secondary sludge (Hämäläinen et al., 2022). Anaerobic digestion of HTC filtrate would be particularly interesting in scenario 3, where the biogas reactor is also needed. The filtrate could be mixed with biosludge to

produce biomethane. Alternative methods for HTC filtrate could make it possible for scenario S1, S2 and S3 to be environmentally more advantageous than the baseline scenario in all impact categories considered.

The production of energy and nutrients carriers in S2, S3 and S4 makes it possible to store the carbon of biosludge instead of emitting it in the atmosphere during combustion, especially in the case of hydrochar for soil amendment. (Yu et al., 2013) also demonstrated the potential of biochar to uptake methane emissions in forest soils, yet in the case of hydrochar, its impact on soils has not been analysed much in literature compared to biochar (Mohammadi et al., 2019a). In this study, the climate impact benefits and improved soil quality of adding hydrochar to the soil in S2 and S3 has not been accounted for. However, according to (Mohammadi et al., 2019a), carbon stabilisation of biosludge hydrochar is a great contributor to climate change mitigation. The results are very sensitive to the amount of stable carbon content in biosludge hydrochar, a data that is not well documented yet, leading to high uncertainty.

This study does not consider the impacts of infrastructures' construction, maintenance and end-of-life. However, their real contribution might impact the results of the study. (Corominas et al., 2020) provides recommendations on the consideration of infrastructures in wastewater treatment LCA studies. Construction phase should be included in the studies, especially for small facilities. Impacts of facilities' maintenance is highly dependent on the choice of technology. End-of-life impacts are often negligible over the operating lifetime of infrastructures and can be neglected. The environmental impacts of construction and maintenance should be allocated evenly across the number of functional units provided. The lifespan of infrastructures in wastewater treatment facilities ranges from 15 years to 100 years according to (Corominas et al., 2020). Concrete structures, equipment and sewer pipes have a respective useful life of approximately 50 years, 15-25 years and 15-100 years. Assuming an annual flow rate of 21,000 t DS biosludge (Saari et al., 2022), the number of functional units provided by infrastructures varies from 315,000 to 2,100,000 FU. In this study, the aim is to compare alternative new treatment technologies with the currently used incineration of biosludge. A more comprehensive study, that would consider the impact of

capital goods (infrastructures and equipment), could therefore include the biomass boiler upgrades over its lifespan, construction of new technologies (HTC, AD, and fermentation process), retrofitting of current infrastructures to match the new treatment methods (increased steam production in S1) and maintenance of all facilities. In this context, scenario S3 that provides the best environmental performance in the present study, would probably results in higher emissions because of the several technologies needed (HTC reactor and belt press, biogas reactor, biogas upgrading facility and digestate belt press).

7. Conclusion

The objective of this study was to evaluate the environmental performance of different treatment methods for pulp and paper biosludge end-of-life, compared to the usually used incineration. The results revealed that hydrothermal carbonization of biosludge for nutrient or energy recovery, as well as combined anaerobic digestion and HTC treatments, show better environmental performances than incineration in four impact categories. Implementation of water recirculation in the HTC reactor might improve further the performance of those three scenarios and would probably make them outperform incineration in all impact categories. Converting biosludge into hydrochar before combustion is particularly interesting from the point of view of climate change as it reduces the emissions related to the biomass boiler by 83%. Separate hydrolysis and fermentation was by far the least preferred option in several impact categories, mainly due to enzyme production. The results for this treatment method are however subject to high uncertainties and very sensitive to enzyme production data. Enzyme production particularly impacts fine particulate matter formation, terrestrial acidification and freshwater eutrophication. Future improvements in enzyme production might help reduce its dosage and environmental impacts. The results of this study were particularly sensitive to the carbon content in the biomass filter for climate change impact category, amount of wastewater going to wastewater treatment plant for human toxicity and marine eutrophication. Terrestrial toxicity results were mainly sensitive to hydrochar, biomethane and bioethanol yields. Considering biogenic emissions deeply changes the results of climate change impact category. Adjusting the contribution factor of biogenic CO₂ to the specific location and type of feedstock is essential to obtain accurate results. It is recommended that additional data should be provided to improve the life cycle assessment of SHF of biosludge. Alternative treatment method for HTC filtrate should also be further investigated. The environmental impact of filtrate recirculation and anaerobic digestion of filtrate should be evaluated and compared with its treatment in wastewater treatment plan. Environmental impact assessment of alternative biosludge handling should be combined with social and economic studies to better assess the sustainability of the different technologies.

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APPENDIX

Appendix 1. Life Cycle Inventory for system 0 – Incineration, excluding boiler and flue gas control (expressed per 1 tonne of dry biosludge).

Belt press - Initial dewatering				
INPUT	Biosludge	33,333	kg	97wt% initial moisture content.
	Electricity	21	kWh	1 kWh per ton of water removed (Heimersson et al., 2014; Stoica et al., 2009).
	Polymer	9	kg	(Mohammadi et al., 2019b)
OUTPUT	Biosludge dried	12,500	kg	92wt% moisture content after dewatering (Saari et al., 2022).
	Water removed	20,833	kg	Directed to WWTP.

Appendix 2. Life Cycle Inventory for system 1 - HTC + Incineration, excluding boiler and flue gas control (expressed per 1 tonne of dry biosludge).

Belt press - Initial dewatering (Same as S0)				
HTC				
INPUT	Biosludge	12,500	kg	
	Electricity	202	kWh	From 120 kWh/t dry BS (Mohammadi et al., 2019a) to 304 kWh/t dry BS (Raatikainen, 2020), excluding the electricity need for dewatering.
	Thermal energy	2,538	kWh	Average value between 0.31 kWh/kg feedstock (Lucian & Fiori, 2017) and 1200 kWh/t dry BS (Mohammadi et al., 2019a).
	Make up water	831	kg	(Saari et al., 2022)
OUTPUT	HTC mix	13,331	kg	(Saari et al., 2022)
	Biogenic CO2 emissions	0.57	kg	(Mannarino et al., 2022)
	CO emissions	0.03	kg	(Mannarino et al., 2022)
	CH4 emissions	156	g	(Mohammadi et al., 2019a)
HTC Belt press				
INPUT	HTC mix	13,331	kg	
	Electricity	20	kWh	(Saari et al., 2022)
OUTPUT	Hydrochar	871	kg	HTC mass yield of 0.82 (Saari et al., 2022; Mannarino et al., 2022; Banks, 2020)
	Filtrate	12,460	kg	Directed to WWTP

Appendix 3. Life Cycle Inventory for system 2 - HTC + Soil amendment, excluding boiler and flue gas control (expressed per 1 tonne of dry biosludge).

Belt press - Initial dewatering (Same as S0)				
HTC (Same as S1)				
HTC Belt press (Same as S1)				
Soil amendment				
INPUT	Hydrochar	871	kg	
EMISSIONS TO SOIL	Cadmium	871	g	(Mohammadi et al., 2019a)
	Chromium	1.2	g	(Mohammadi et al., 2019a)
	Copper	8.3	g	(Mohammadi et al., 2019a)
	Nickel	6.4	g	(Mohammadi et al., 2019a)
	Lead	2.5	g	(Mohammadi et al., 2019a)
	Zinc	2.4	g	(Mohammadi et al., 2019a)
FERTILIZER USE AVOIDED	N fertilizer (NH ₄ NO ₃)	20.1	kg	(Mohammadi et al., 2019a)
	P fertilizer (P ₂ O ₅)	6.53	kg	(Mohammadi et al., 2019a)
	K fertilizer (K ₂ O)	0.87	kg	(Mohammadi et al., 2019a)
	Mg fertilizer (MgO)	0.80	kg	(Mohammadi et al., 2019a)
	Ca fertilizer (CaCO ₃)	0.13	kg	(Mohammadi et al., 2019a)
TRANSPORT	Distance to forest site	100	km	(Mohammadi et al., 2019a)

Appendix 4. Life Cycle Inventory for system 3 - AD + HTC + Soil amendment, excluding boiler and flue gas control (expressed per 1 tonne of dry biosludge).

Belt press - Initial dewatering (Same as S0)				
AD + Biogas upgrading				
INPUT	Biosludge	12,500	kg	
	Electricity	53	kWh	Average value from (Li & Feng, 2018; Mainardis et al., 2021; Mohammadi et al., 2019b)
	Thermal energy	370	kWh	(Heimersson et al., 2017)
OUTPUT	Biomethane	1025	kWh	(Mainardis et al., 2021; Mannarino et al., 2022; Mohammadi et al., 2019b, Kinnunen et al., 2015). 1% of the production is leaked (Heimersson et al., 2017)
	Digestate	570	kg dry solids	(Heimersson et al., 2017)
	CH4 leakage	0.73	kg	1% of biomethane production (Heimersson et al., 2017)
	Nitrous oxide	0.02	kg	(Heimersson et al., 2017)
	Biogenic CO2	113	Kg	Calculated according to (Møller et al., 2009)
HTC of digestate - Flows are calculated based on dry solids input ratio of 0.57 compared to S1				
FEEDSTOCK	Digestate	570	kg dry solids	
HTC Belt press - Flows are calculated based on dry solids input ratio of 0.57 compared to S1				
FEEDSTOCK	HTC mix	7,598	kg	
Soil amendment - Flows are calculated based on dry solids input ratio of 0.57 compared to S2				
FEEDSTOCK	Hydrochar	566	kg	
TRANSPORT	Distance to forest site	100	km	(Mohammadi et al., 2019a)

Appendix 5. Life Cycle Inventory for system 4 - Fermentation, excluding boiler and flue gas control (expressed per 1 tonne of dry biosludge).

Belt press - Initial dewatering (Same as S0)			
Enzymatic hydrolysis			
INPUT	Biosludge	12,500 kg	
	Steam	896 kg	Total steam demand for the whole process of separate hydrolysis and fermentation (Chen et al., 2014)
	Enzymes	33 kg	(Chen et al., 2014)
	Sulphuric acid	259 kg	(Chen et al., 2014)
OUTPUT	Hydrolyzed biosludge	4,900 kg	(Chen et al., 2014)
Fermentation			
INPUT	Hydrolyzed biosludge	4,900 kg	
	Yeast	3 kg	(Dunn et al., 2012)
OUTPUT	Fermented biosludge	4,717 kg	(Chen et al., 2014)
Distillation			
INPUT	Fermented biosludge	4,717 kg	
	Steam	0 kg	Included in the amount of steam needed for enzymatic hydrolysis
OUTPUT	Bioethanol	173 kg	(Chen et al., 2014)
	Residues	4,259 kg	(Chen et al., 2014)
Belt press 2			
INPUT	Residues	4,259 kg	
	Electricity	3 kWh	1 kWh per ton of water removed (Heimersson et al., 2014; Stoica et al., 2009)
OUTPUT	Dried residues	989 kg	30% dry solids (Chen et al., 2014)
	Water removed	3270 kg	Directed to WWTP
Residues to waste incineration			
INPUT	Dried residues	989 kg	LHV assumed to be 3.78 MJ/kg based on biosludge LHV and dried residues moisture content.
OUTPUT	Steam	669 kWh	Gabi database
	Electricity	16 kWh	Gabi database

Appendix 6. Life Cycle Inventory for the boiler and flue gas control of every scenario (expressed per 1 tonne of dry biosludge).

INPUTS	Unit	S0	S1	S2	S3	S4	References
Feedstock							
Biosludge	kg	12,500	0	0	0	0	
Hydrochar	kg	0	871	0	0	0	
Bark	kg	40,096	40,096	40,096	40,096	40,096	(Saari et al., 2022)
Fines	kg	4,005	4,005	4,005	4,005	4,005	(Saari et al., 2022)
Operation		138,925	146,181	141,796	141,796	141,796	
Feed water	kg	638	619	611	611	611	Internal calculations.
Quarry sand	kg	12,500	0	0	0	0	(Heimersson et al., 2014)
Chemicals for flue gas treatment							
Sodium hydroxide	kg	779	755	745	745	745	(Mohammadi et al., 2019b)
Sulphuric acid	kg	28	27	26	26	26	(Mohammadi et al., 2019b)
Sodium chloride	kg	14	13	13	13	13	(Mohammadi et al., 2019b)
Hydrochloric acid	kg	46	45	44	44	44	(Mohammadi et al., 2019b)
Calcium chloride	kg	15	15	14	14	14	(Mohammadi et al., 2019b)
Sodium persulfate	kg	15	14	14	14	14	(Mohammadi et al., 2019b)
Ammonia	kg	11.0	9.8	9.4	9.4	9.4	(Mohammadi et al. 2019b)
OUTPUTS	Unit	S0	S1	S2	S3	S4	
Energy							
Heat for self-consumption*	kWh	0	2,538	2,538	1,816	1,736	Internal calculations.
Heat to sell	kWh	59,441	59,568	58,995	59,716	62,525	Internal calculations.
Electricity for self-consumption*	kWh	21	242	242	200	30	Internal calculations.
Electricity to sell	kWh	34,392	35,713	35,382	35,424	35,660	Internal calculations.
Solid waste							
Ash	kg	98	71	62	62	62	(Mohammadi et al., 2019)
Flue gas emissions							
Dry flue gas	kg	192,851	188,156	185,769	185,769	185,769	
CO2	kg	44,924	43,915	43,405	43,405	43,405	(EPA, 1995)
NO2	g	21	19	18	18	18	(Mohammadi et al. 2019b)
SO2	g	1,244	882	744	744	744	2% of S content in fuel (Havukainen et al. 2022)
NH3	g	1.20	1.07	1.02	1.02	1.02	(Mohammadi et al. 2019b)
N2O	kg	25.5	20.0	19.9	19.9	19.9	
Heavy metals							
Cd	mg	3.8	15.5	0	0	0	Transfer coefficients of heavy metals from fuel to exhaust gas is taken from (Heimersson et al. 2014)
Cr	mg	42.9	35.9	0	0	0	
Cu	mg	4.0	5.7	0	0	0	
Ni	mg	24.7	21.2	0	0	0	
Pb	mg	5.0	8.2	0	0	0	
Zn	mg	575	806	0	0	0	

*Heat and electricity consumption within the system boundaries

Appendix 7. LCIA results and contribution analysis.

	Total	Initial DW	Boiler & FGT	HTC	AD	SFH	Soil amendment	Transport	Electricity from the grid	Heat production	Fertilizer production	Natural gas production	Gasoline production
Climate change [kg CO2 eq.]													
S0	53,516	< 1%	99%	-	-	-	-	-	< 1%	< 1%	< 1%	< 1%	< 1%
S1	50,584	< 1%	100%	< 1%	-	-	-	-	-	< 1%	< 1%	< 1%	< 1%
S2	50,145	< 1%	99%	< 1%	-	-	-	< 1%	< 1%	< 1%	-	< 1%	< 1%
S3	50,232	< 1%	99%	< 1%	< 1%	-	-	< 1%	< 1%	< 1%	< 1%	-	< 1%
S4	50,944	< 1%	98%	-	-	2%	-	-	< 1%	-	< 1%	< 1%	-
Fine particulate matter formation [kg PM2.5 eq.]													
S0	0.79	< 1%	34%	-	-	-	-	-	24%	31%	< 1%	1%	8%
S1	0.51	< 1%	52%	< 1%	-	-	-	-	-	32%	< 1%	2%	13%
S2	0.72	< 1%	36%	< 1%	-	-	-	2%	10%	40%	-	2%	9%
S3	0.62	< 1%	42%	< 1%	-	-	-	1%	11%	34%	< 1%	-	10%
S4	2.36	< 1%	11%	-	-	86%	-	-	2%	-	< 1%	< 1%	-
Terrestrial acidification [kg SO2 eq.]													
S0	2.49	< 1%	35%	-	-	-	-	-	24%	30%	< 1%	1%	9%
S1	1.64	< 1%	52%	< 1%	-	-	-	-	-	31%	< 1%	2%	13%
S2	2.27	< 1%	37%	< 1%	-	-	-	2%	10%	39%	-	1%	10%
S3	1.96	< 1%	43%	< 1%	-	-	-	1%	11%	33%	< 1%	-	11%
S4	8.05	< 1%	10%	-	-	87%	-	-	2%	-	< 1%	< 1%	-
Freshwater eutrophication [kg P eq.]													
S0	0.023	55%	35%	-	-	-	-	-	2%	< 1%	< 1%	< 1%	8%
S1	0.031	42%	27%	25%	-	-	-	-	-	< 1%	< 1%	< 1%	6%
S2	0.031	42%	27%	25%	-	-	-	< 1%	< 1%	< 1%	-	< 1%	6%
S3	0.028	47%	30%	16%	-	-	-	< 1%	< 1%	< 1%	< 1%	-	7%
S4	0.207	6%	4%	-	-	90%	-	-	< 1%	-	< 1%	< 1%	-
Terrestrial ecotoxicity [kg 1.4-DB eq.]													
S0	395	< 1%	45%	-	-	-	-	-	21%	12%	< 1%	9%	12%
S1	341	< 1%	65%	< 1%	-	-	-	-	-	9%	< 1%	11%	14%
S2	245	1%	28%	< 1%	-	-	< 1%	< 1%	13%	22%	-	15%	20%
S3	190	1%	36%	< 1%	-	-	< 1%	< 1%	16%	20%	< 1%	-	26%
S4	167	2%	41%	-	-	23%	-	-	12%	-	< 1%	22%	-
Human toxicity. cancer [kg 1.4-DB eq.]													
S0	2.26	47%	35%	-	-	-	-	-	10%	3%	< 1%	< 1%	5%
S1	2.67	40%	30%	24%	-	-	-	-	-	2%	< 1%	< 1%	4%
S2	2.91	37%	27%	22%	-	-	5%	< 1%	3%	3%	-	< 1%	4%
S3	2.54	42%	31%	14%	-	-	3%	< 1%	3%	2%	< 1%	-	4%
S4	2.00	53%	39%	-	-	5%	-	-	3%	-	< 1%	< 1%	-
Marine eutrophication [kg N eq.]													
S0	0.070	35%	39%	-	-	-	-	-	6%	< 1%	< 1%	< 1%	19%
S1	0.081	30%	34%	18%	-	-	-	-	-	< 1%	< 1%	< 1%	16%
S2	0.082	30%	33%	18%	-	-	-	< 1%	2%	< 1%	-	< 1%	16%
S3	0.075	33%	36%	11%	-	-	-	< 1%	2%	< 1%	< 1%	-	18%
S4	0.056	44%	49%	-	-	4%	-	-	2%	-	1%	< 1%	-

AD: anaerobic digestion; DW: dewatering; FGT: flue gas treatment; HTC: hydrothermal carbonization; SHF: separate hydrolysis and fermentation.