Master's thesis

NTNU
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Department of Energy and Process Engineering

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Life cycle assessment of sewage sludge treatment methods for negative emissions and abatement of hazardous contaminants

Master's thesis in Energy and Environmental Engineering
Supervisor: Francesco Cherubini
Co-supervisor: Marjorie Morales, Otávio Cavalett
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Preface

This thesis concludes my Master of Science in Energy and Environmental Engineering at the Norwegian University of Science and Technology (NTNU) and was written in collaboration with the Industrial Ecology Programme at NTNU. It continues the work of my project thesis from the autumn semester of 2020, "Thermal treatments of sewage sludge for abatement of hazardous substances and biochar production for negative emissions", where the fate of hazardous substances in sewage sludge treatment and the negative emission potential of sewage sludge were studied. In this thesis, the complete life cycle impacts of the treatment methods and the final disposal of sewage sludge is studied further in order to capture the environmental impacts if these treatment scenarios from a life cycle perspective.

I would like to thank my supervisor, Francesco Cherubini, and my co-supervisors, Marjorie Morales and Otávio Cavalett, for all their guidance and valuable feedback. Working with the specialization project and thesis has been a long learning process, and the end result would not have been possible without your continuous help and motivation. I also want to express my gratitude to my friends and family for supporting me during my studies.

11.06.2021
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Marit Salome Rognan

Abstract

Sewage sludge (SS) treatment and disposal do not take full advantage of the energy and nutrients in SS, and often leads to emissions of greenhouse gases (GHGs) and pollutants. This study explores treatment methods involving anaerobic digestion (AD) and pyrolysis for the conversion of SS into the energy-rich products biogas, bio-oil, biochar and syngas. The biochar, if applied to soil, can be used to sequester carbon and recycle nutrients from the SS. It may also be possible to use as a solid fuel to decarbonize industries. To fully include the effect of pyrolysis of SS, the fate of several hazardous organic pollutants (HOCs) and heavy metals (HMs) is carefully assessed in each scenario.

A comparative life cycle assessment (LCA) was performed using data from the literature adapted to Norwegian conditions. The main objective was to identify and compare the environmental impacts of alternative SS management scenarios using pyrolysis with a reference case for conventional Norwegian SS treatment and disposal. The results show that the largest negative emissions are achieved when using slow pyrolysis at 300°C to produce biochar that can substitute coal combined with carbon capture and storage (CCS), where up to 62% of the carbon in SS could be captured. However, to minimize the climate impacts, it was found that that a combination of AD and pyrolysis at 500°C followed by soil application of biochar is the preferred scenario, but here only 10% of the carbon from the initial SS ends up sequestered. It was also found that slow pyrolysis at 500°C is a promising method to reduce the bioavailability of HMs and destroy HOCs in the SS. Moreover, the LCA showed that HMs, not HOCs, are the main contributor to human toxicity and freshwater ecotoxicity from soil application of Norwegian SS.

Due to the additional bioenergy products from pyrolysis and reduced GHG emissions from soil application, the scenarios with pyrolysis came out better in terms of their life cycle impacts. That being said, additional research on the fate of HOCs and HMs during SS pyrolysis is recommended before agricultural application of SS biochar. The accumulation of HMs in the biochar poses the main challenge for its use in agriculture, but it was found the bioavailability of HMs was reduced significantly during pyrolysis.

Sammendrag

Behandling og bruk av avløpsslam utnytter generelt ikke all energien og næringsstoffene som finnes i slammet, og fører ofte til utslipp av klimagasser og forurensende stoffer. Denne studien studerer behandlingsmetoder som involverer anaerob stabilisering (AD) og pyrolyse for å omdanne slammet til biogass, bioolje, biokull og syngass. Hvis biokullet blandes i jorda vil det bidra til negative karbonutslipp og resirkulering av næringsstoffer. Det kan også være mulig å bruke biokullet for å avkarbonisere industri. For å best mulig inkludere effekten av pyrolyse av slam, blir skjebnen til noen særlig prioriterte organiske miljøgifter og tungmetaller vurdert nøye i hvert scenario.

En livssyklusvurdering (LCA) ble utført ved hjelp av data fra litteraturen tilpasset norske forhold. Hovedmålet var å identifisere og sammenligne miljøpåvirkningen av alternative behandlingsmetoder av slam hvor pyrolyse benyttes, med et referansescenario for konvensjonell behandling og bruk av slam i Norge. Resultatene viser at karbonfangstpotentialet er høyest i scenarioet der langsom pyrolyse ved 300°C ble brukt for å produsere biokull til å erstatte kull i kombinasjon med karbonfangst og lagring (CCS). Opptil 62 % av karbonet i slammet kunne fanges her. For å minimere klimaeffektene ble det imidlertid funnet at en kombinasjon av AD og pyrolyse ved 500°C, etterfulgt av jordpåføring av biokull, er det foretrukne scenariet. Her er det riktignok bare 10 % av karbonet fra slammet som ender opp permanent lagret. Det ble også funnet at langsom pyrolyse ved 500°C virker lovende for å redusere biotilgjengeligheten til tungmetaller og å ødelegge organiske miljøgifter i slam. Videre viste resultatene at det er tungmetaller som er den største bidragsyteren til negativ påvirkning på menneskelig helse og økosystemer ved slamdisponering på land, sammenlignet med de andre særlig prioriterte miljøgiftene.

På grunn av ekstra bioenergiprodukter fra pyrolyse og reduserte klimagassutslipp ved disponering av biokull, hadde scenariene med pyrolyse reduserte livssykluspåvirkninger sammenlignet med referansescenariet. Når det er sagt, anbefales ytterligere forskning på skjebnen til miljøgifter og tungmetaller under pyrolyse, før biokull fra slam brukes i landbruk. Akkumulering av tungmetaller i biokull er en utfordring for bruk i landbruk, men det ble funnet at biotilgjengeligheten deres ble redusert betydelig under pyrolyse.

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Abbreviations

AD = Anaerobic digestion
AHTN = Tonalide
BDE-209 = DecaBDE
BPA = Bisphenol A
CCS = Carbon capture and storage
DEHP = Di-(2-ethylhexyl) phthalate
DM = Dry matter
EOFP = Ozone formation potential, Terrestrial ecosystems
FDP = Fossil resource scarcity potential
FEP = Freshwater eutrophication potential
FETP = Freshwater ecotoxicity potential
GHG = Greenhouse gas
GWP = Global warming potential
HHCB = Galaxolide
HM = Heavy metal
HOC = Hazardous organic contaminant
HOFP = Ozone formation potential, Human health
HTPc = Human toxicity potential, cancer
HTPnc = Human toxicity potential, non-cancer
IRP = Ionizing radiation potential
LAS = Linear alkylbenzene sulfonate
LCA = Life cycle assessment
LOP = Land use potential
MC = Moisture content
MDP = Mineral resource scarcity potential
MEP = Marine eutrophication potential
METP = Marine ecotoxicity potential
NET = Negative emission technology
NG = Natural gas
NP = Nonylphenol
NPE = Nonylphenol ethoxylate
ODP = Stratospheric ozone depletion potential
OTNE = Iso-E-Super
PAH = Polycyclic aromatic hydrocarbon
PCB = Polychlorinated biphenyls
PCDD/F = Polychlorinated dibenzo-p-dioxin and -furan
PFOA = Perfluorooctanoic acid
PFOS = Perfluorooctane sulfonate
PMPF = Fine particulate matter formation potential
SCCP = Short-chained chlorinated paraffin
SMR = Steam methane reforming
SS = Sewage sludge
TAP = Terrestrial acidification potential
TETP = Terrestrial ecotoxicity potential
VM = Volatile matter
WCP = Water consumption potential
WWTP = Wastewater treatment plant

1 Introduction

1.1 Background and motivation

Mitigating climate change is becoming increasingly challenging as anthropogenic carbon dioxide (CO_2) emissions continue to increase [1]. If we are to reach the Paris agreement of maintaining the global average temperature to well below 2 °C above preindustrial levels, it is clear that we need to implement negative emission technologies (NETs) and reduce emissions of greenhouse gases (GHGs) [2].

Sewage sludge (SS), the residue from wastewater treatment processes, is rich in organic matter and has the potential to be used as both a renewable energy source or a carbon sink. SS contains many compounds of agricultural value, like phosphorus (P) or nitrogen (N), as well as many pollutants and pathogens [3]. Instead of being regarded as inferior waste, a shift in public perception is needed to incorporate SS as part of a circular economy. It is estimated that SS contains enough nutrients to replace 25% of the N and 15% of P currently used in synthetic fertilizers in agriculture [4]. Moreover, its carbon content offers a negative emission potential if sequestered in a stable form in soil [4].

Improving SS treatment and disposal would contribute to six of the 17 SDGs, as shown in figure 1.1. Improper treatment and disposal of SS is not only a loss of a valuable resource - its potential for eutrophication and its toxicity can cause long-term damage to ecosystems and humans due to the high content of nutrients, hazardous organic contaminants (HOCs) and heavy metals (HMs). Because of the accumulation of HOCs and HMs in SS, some countries have completely banned it to be spread on land. However, the alternative disposal method is often incineration or landfilling. To achieve a more circular economy, the optimal waste management involves high recycling, low incineration and low landfilling [5]. In addition, landfilling and incineration of SS contribute to air pollution and emissions of GHGs [6, 7]. There is therefore a need to identify and implement SS treatment options that can effectively reduce the toxicity of the SS, while simultaneously allowing the extraction of valuable energy, conservation of mass and nutrients and utilizing its negative emission potential.

Figure 1.1: The Sustainable Development Goals (SDGs) impacted by wastewater and sewage sludge management.

[8]

Pyrolysis is a promising technique that can effectively convert biomass into bio-oil, syngas and biochar. During pyrolysis, biomass is thermally decomposed under inert conditions and elevated temperatures [9]. The bio-oil and syngas can be burned for energy purposes, and the carbon-rich residue, biochar, can be applied to soil as a soil amendment and carbon sink [10, 11]. Because pyrolysis is also effective in immobilizing HMs and potentially destroying many HOCs, the biochar produced from SS pyrolysis could be a more attractive soil amendment option than SS is today. Furthermore, because the carbon is locked in a stable form in the biochar, the land application of biochar enables long-term carbon sequestration [10]. Given the biochar's high energy content and coal-like properties, it could potentially be used as a substitute to coal to decarbonize industries [9, 12], which, combined with carbon capture and storage (CCS), would also contribute to negative emissions.

Carbon sequestration in soil can help mitigate climate change as it represents a large and long-term carbon sink [10]. Generally, biochar application to land is one of the most affordable negative emission technologies (NETs) with few disadvantages and trade-offs [13, 14]. Figure 1.2 depicts how biochar addition to soil acts as negative carbon emissions by sequestering the carbon from the atmosphere in the land. The biochar can be produced from a wide range of feedstocks, both plant-based and animal-based. That being said, access to biomass feedstocks that does not increase pressure on ecosystems is key, and the use of existing residues and waste should therefore be prioritized. Experiments show that the carbon in biochar has a very high stability after soil application also when applied on soils in Norway [15].

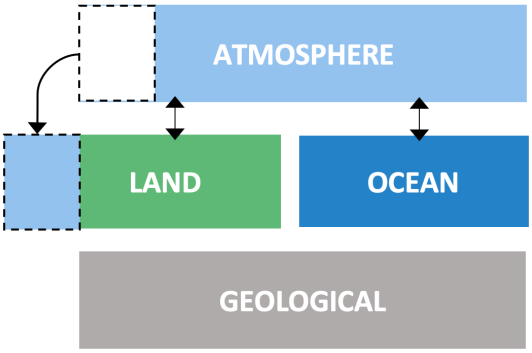


Figure 1.2: Flow of carbon for biochar addition to soil, adapted from Smith et al. [14]

The co-benefits of land application of biochar depend on a multitude of factors. Several studies have found that it is capable of improving yield and soil quality and even reduce soil GHG emissions [13]. SS, when applied to land after traditional treatment, releases GHGs like nitrous oxide (N2O) and will eventually release its carbon back to the atmosphere [6]. If SS-derived biochar is used instead, the majority of the carbon will remain in the soil on a centennial scale, and the co-benefits could give reduced GHG emissions from the soil [16, 17].

National statistics for Norway show an average SS generation in the past five years of 114 000 tons dry matter (DM) annually [18]. Figure 1.3 shows some of the most common SS treatment pathways for SS in Norway. The main purpose of the treatment is to stabilize and sanitize the SS, and in addition reduce the water content for further use or disposal. Some methods are also able to convert part of the organic matter into energy products, such as the method anaerobic digestion (AD). During AD, organic matter is broken down in an oxygen-free environment to produce a methane rich biogas. It is the only energy-positive technology widely used in wastewater and sewage treatment [19], however, it can only convert around 40-50% of the organic matter into biogas [3]. AD can also increase biodegradation of HOCs, however for many HOCs it is likely negligible [20].

By combining AD and pyrolysis, all the DM in the SS can be converted to biogas, bio-oil, syngas and biochar. The biogas is a more valuable energy product than the syngas and bio-oil from SS because it has little impurities and a very high methane content. Several studies have found that combination of AD and pyrolysis has lower life cycle impacts than using only pyrolysis [21, 22]. On the other hand, pretreating the SS with AD before pyrolysis results in less of the carbon in the SS ending up in the biochar, which in turn reduces the negative emission potential. There is therefore potentially a trade-off between the carbon sequestration and the production of renewable fuels.

As shown in figure 1.4, more than 80% of the SS from public wastewater treatment plants in Norway is recycled back to the soil through agricultural application, application on greenery or delivered to a soil producer [18]. Due the presence of contaminants in SS there are strict restrictions on its use in agriculture in Norway on the maximum allowable application rate and interval between applications, making it a soil product of low value to farmers. This is even stricter in other countries where little to no SS is

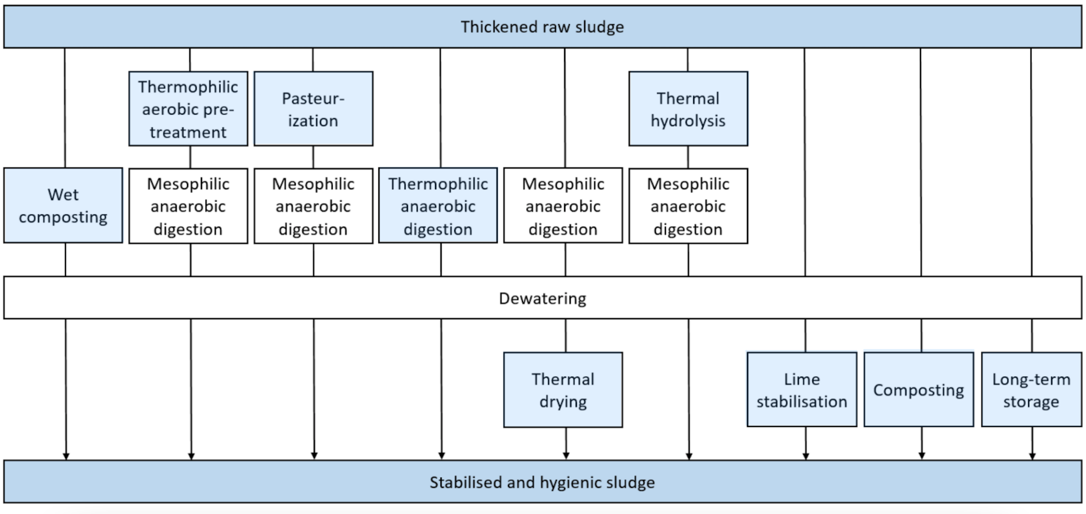


Figure 1.3: Overview of common SS treatment processes applied in Norway. Sanitizing steps are in light-blue boxes.

[20]

recycled back to the soil. One example is Sweden, where only one fifth of the produced SS is used on land [23], which furthermore cannot be agricultural land.

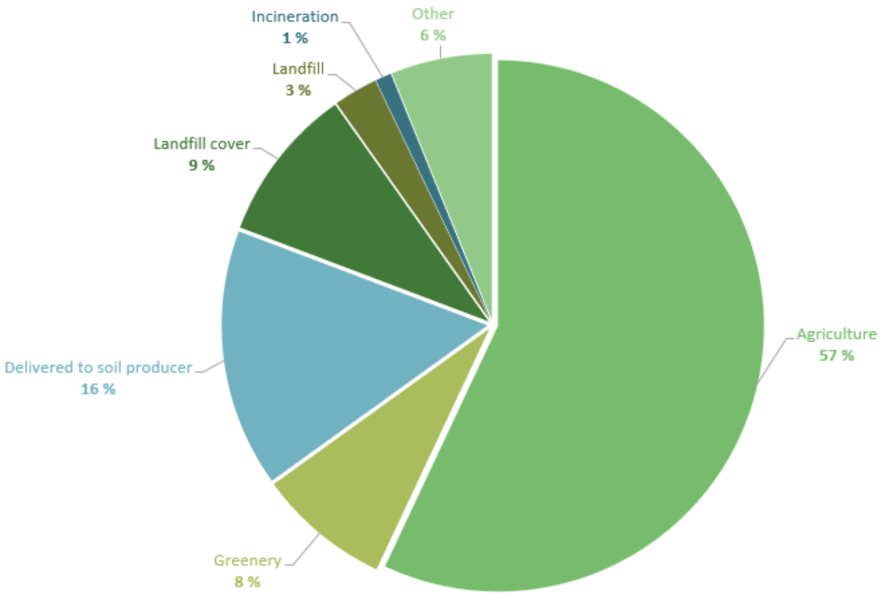


Figure 1.4: Disposal (% of total) of SS in Norway from year 2015-2019.

1.2 Aim of the study

In this study, a life cycle assessment (LCA) of SS treatment methods using pyrolysis for negative emissions and abatement of hazardous compounds will be performed using the available literature and the LCA software SimaPro (9.1.1.1). Whenever possible, the study uses data relevant for Norway and Norwegian SS management. **The aim of the study is to compare the environmental impacts of alternative SS treatment scenarios where pyrolysis is used, with a baseline scenario of a commonly used SS treatment and disposal method in Norway**. In addition to the total life cycle impacts, the study's secondary aims are to assess the potential destruction of HOCs during AD and pyrolysis, and the negative emission potential the treatment pathways can offer.

While several LCA studies have already been done on the treatment of SS using variations of AD and pyrolysis, they do not quantitatively take into account the fate of HOCs and their potential destruction during SS pyrolysis. The contributions of HOCs to the life cycle impacts from SS disposal therefore remain unknown. In this study, a selection of priority HOCs in Norwegian SS are included, as well as the regulated HMs for SS soil application. Their emissions from sludge treatment and disposal will be quantified based on the available literature. The next section, Methods, will present the treatment and disposal scenarios that will be analyzed in this thesis, as well as the reasoning behind this choice of process conditions and how their life cycle inventory and impacts will be determined.

2 Methods

2.1 Scenario description

The scenarios to be compared with the reference case are illustrated in figure 2.2 and summarized in table 2.1. For simplicity, the reference case of SS treatment is chosen to also be mesophilic AD followed by dewatering and thermal drying, as shown in 2.1, but as previously mentioned there are many methods in use.

Table 2.1: The scenarios to be assessed in the LCA.

Scenario	Treatment pathway	Product uses
Reference case	Mesophilic AD, dewatering and drying	Biogas used combined heat and power (CHP) system on-site and biosolids spread on land
1: AD+PY500, CHP	Mesophilic AD, dewatering, drying and slow pyrolysis at 500°C	Biochar spread on agricultural land and the co-products used in a CHP system on-site
2: AD+PY500, upgr.	Mesophilic AD, dewatering, drying and slow pyrolysis at 500°C	Biochar spread on agricultural land, biogas is upgraded to substitute NG, bio-oil is upgraded to substitute diesel and the syngas is used in a CHP system on-site
3: AD+PY300, CHP	Mesophilic AD, dewatering, drying and slow pyrolysis at 300°C	Biochar is used in industry to replace coal combined with CCS and the co-products go to a CHP system on-site
4: PY500, CHP	Dewatering, drying and slow pyrolysis at 500°C	Biochar spread on agricultural land and and co-products are used in a CHP on-site
5: PY300, CHP	Dewatering, drying and slow pyrolysis at 300°C	Biochar used in industry to replace coal combined with CCS and co-products used in CHP on-site

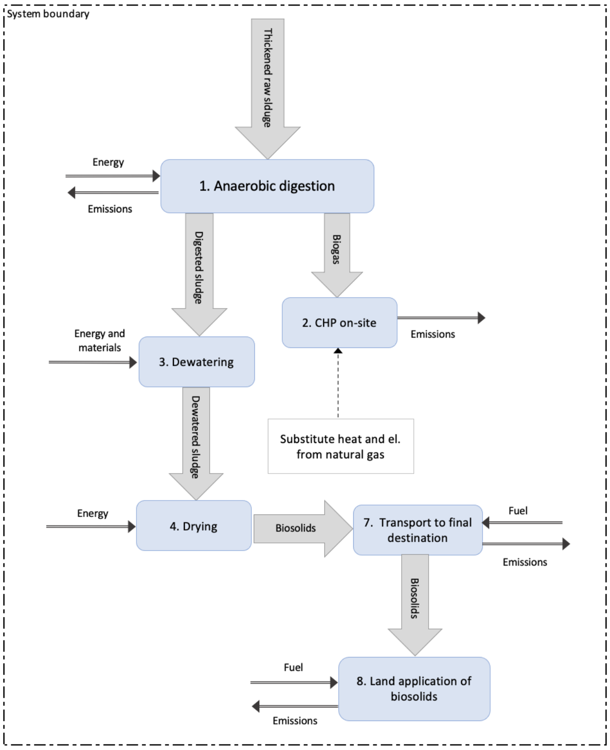
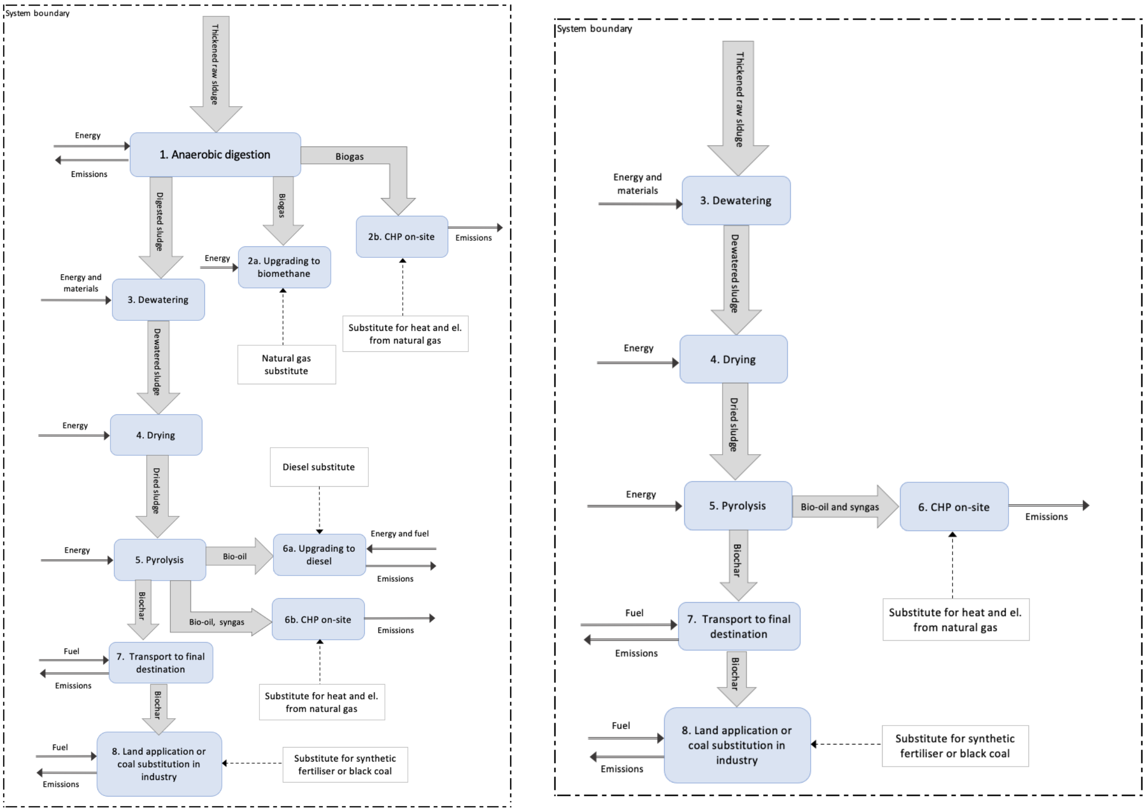


Figure 2.1: LCA system boundaries for the reference case of Norwegian SS treatment and disposal.



(a) LCA system boundaries for the systems with AD and pyrolysis (scenarios 1-3).

(b) LCA system boundaries for the systems with only pyrolysis (scenarios 4-5).

Figure 2.2: LCA system boundaries scenarios 1-5.

2.2 Determining sewage sludge composition and pollutants

The composition of SS can vary strongly depending on its source, such as whether it originates from a municipal wastewater treatment plant (WWTP) or an industrial WWTP, as well as local and national variations in the wastewater. SS from conventional WWTPs is derived from either primary, secondary and tertiary treatment processes. Primary SS is produced following a primary treatment of wastewater, which consists of physical or chemical treatments to remove matter in suspension [3]. The secondary SS is generated by decomposers that attempt to break down remaining organic materials in wastewater after primary treatment. Lastly, tertiary SS is generated when carrying out tertiary treatment with the goal of removing remaining unwanted nutrients, like N and P. These treatments are only necessary if a high level of depollution is required and its use varies between different countries [3].

2.2.1 General composition

Due to the lack of a country specific SS composition for Norway, a general SS composition of mixed SS (mix of primary and secondary SS) in the EU was chosen for this study to stay to consistent with some of the main references. The composition is shown in table 2.2. It was documented by the European Commission in 2001 based on data from member countries and might therefore be less representative today.

Table 2.2: General SS composition for mixed SS.

Proximate analysis	
Moisture content (% wt.)	95.0
Volatile solids (% DM)	72.0
Fixed carbon (% DM)	8.0
Ash (% DM)	27.2

Ultimate analysis (% DM)	
C	36.72
H	5.33
N	5.11
Cl	0.80
S	1.08
O	23.76

[3]

2.2.2 Organic contaminants

SS contains many organic contaminants, pathogens and HMs that could pose a risk for humans, animals and ecosystems if released back into water or soil. While some of these biodegrade or volatilize during SS treatment, many are resistant to biodegradation or volatilizing and are not removed significantly during treatment. They are also strongly sorbed onto the solids in the SS because of their lipophilic properties, making mechanical methods ineffective for their removal as well.

Unlike for the general SS composition, measurements are done regularly for concentrations of HOCs and HMs in Norwegian SS. Hundreds of different organic pollutants and medicinal residues are among these compounds under surveillance [24], but for this study only a selection of them will be included.

Based on several reports and studies on Norwegian SS toxins as well as a Danish study on priority pollutants in SS, 14 different HOCs were chosen to be included in this study. These studies aimed to determine which organic contaminants pose the highest risk for using SS derived products as fertilizer and soil amendment products. This thesis will include the pollutants selected by the Norwegian Environment Agency to be evaluated by NIBIO based on their risk when present in soil products. These HOCs are persistent and bioaccumulative, have serious long-time effects, effects on reproduction and genetics or are strongly toxic to the environment [20]. The Danish study determined priority pollutants based on a similar hazard identification and assessment as well as an expert judgement [23]. The resulting priority HOCs, where concentrations were available for Norwegian SS, are shown in table 2.3. The measured concentrations of the HOCs are given in table B1 in the Appendix.

Table 2.3: Priority SS organic contaminants chosen for this study.

Group	Selected compound(s)
Endocrine disruptors	Nonylphenol (NP) Nonylphenol ethoxylates (NPE) Bisphenol A (BPA)
Phtalates and plasticizers	Di-(2-ethylhexyl) phthalate (DEHP)
Flame retardants	DecaBDE (BDE-209)
Polycyclic aromatic hydrocarbons (PAHs)*	PAH_{16}
Pesticides	Triclosan
Perfluorinated alkyl acids (PFAS)	PFOS PFOA
Chlorinated paraffins	Short-chained chlorinated paraffins (SCCP)
Synthetic musks	HHCB AHTN OTNE
Polychlorinated biphenyls (PCB)	PCB 7
Anionic surfactants	Linear alkylbenzene sulfonates (LAS)

*The individual compounds of PAHs that are measured might vary between studies as it is not always specified. PAH_{16} is used when measuring for PAHs in Norwegian SS [25].

2.2.3 Heavy metals

A strictly regulated aspect of organic fertilizer products is their concentration of HMs. While there are currently no maximum limits for organic pollutants in SS-derived fertilizers in Norway, there are maximum limits for the following HMs [26]:

- Lead (Pb),
- Zink (Zn),
- Nickel (Ni),
- Cadmium (Cd),
- Copper (Cu),
- Chromium (Cr),
- Mercury (Hg)

The concentrations of these HMs determine if, where and how often the fertilizer can be applied. If SS is used for biochar production, the HMs will largely accumulate in the biochar and that is a concern for its reuse on land, particularly in agriculture. The average measured concentrations in Norwegian SS of these regulated HMs are given in table B3 in the Appendix.

More importantly however, when considering the toxicity the HMs pose, is their bioavailability or leachability. The bioavailable fraction of a HM represents the fraction of a compound/element that is accessible for absorption for plants [27]. In a recent study by Wang et al. [28], it was found that although the concentration of HMs increased in the biochar compared to the SS feed, the pyrolysis inhibited their ecological risks by transforming them from mobile to stable fractions. Several other studies also confirm this effect of SS pyrolysis on HMs [27,29,30]. Therefore, to more accurately represent the ecotoxicity of the HMs in this study, their estimated bioavailability when applied to land is used to quantify the HM emissions to soil. Studies with similar process conditions for biosolids or biochar production are used as a basis for determining the bioavailable fraction of the HMs in the biosolids and biochar applied to land.

2.3 Research on treatment and disposal methods

2.3.1 Anaerobic digestion

During AD of SS, micro-organisms break down part of the organic matter and produce an energy-rich biogas containing mainly methane ($CH_{4)}$ and CO_2. It is therefore a commonly used method for SS treatment and overall performs well in reducing SS volume/weight, pathogens and pharmaceuticals, as well as global warming potential (GWP) reduction [22]. The process can take place in ambient temperature (psychrophilic AD), 30–38°C (mesophilic AD) or 50–57°C (thermophilic AD). Mesophilic AD remains dominant in practice because of its acceptable energy consumption, reliable process operation and favorable process performances [31]. There also exists a lot of data on the biodegradation of HOCs during mesophilic AD. It is therefore chosen to be included in this study.

Given the mesophilic process conditions, the fraction of organic matter that is transformed to biogas is assumed to be 56% of the volatile solids (VS) in the SS [32]. To estimate the heat and electricity demand of the mesophilic AD, it was necessary to also assume the size of the digester. IVAR wastewater treatment plant is one of the treatment facilities in Norway using the treatment pathway consisting of AD, dewatering and drying. They have three digester tanks at 3500 m^3 each, and with a total retention time of 15 days and solids content of 5% this gives a daily solids loading of

$$3 \cdot 3500\, m^3 \cdot \frac{1\,\text{ton}}{m^3} \cdot 5\% DM \div 15\,\text{days} = 35\ \text{tons DM} \tag{1}$$

With a daily loading of 35 tons DM, the empirical equations for predicting energy consumption based on facility capacity in [33], gives a speficic heat and electricity consumption for mesophilic AD of 1100 MJ/ton DM and 125 kWh/ton DM, respectively.

Table 2.4: Summary of the process parameters in the mesophilic AD.

Parameter	Unit	Value	Reference
Temperature	°C	35	[32]
Retention time	days	15	[32]
Volatile solids (VS) destruction	% VS	56	[32]
Heat consumption[a]	MJ/ton DM	1100	[33]
Electricity consumption[a]	kWh/ton DM	125	[33]
Biogas production	m^3/kg VS destroyed	1	[32]
CH_4 in biogas	% vol.	65	[32]
CO_2 in biogas	% vol.	35	[32]

[a] Using IVAR wastewater treatment plant as reference for the daily load [34].

Several studies have been done on the fate of HOCs during mesophilic AD, though with somewhat conflicting results. While some studies find that a significant fraction of many HOCs is removed or biodegraded, others find that only a negligible amount is removed. It is also worth noting that these studies are done in lab-scale digesters, so the realistic removal in large-scale digesters might be different than the estimates used here. Where several removal rates have been found for the same substance with similar process conditions and feedstock, the average of those values is used. It is also worth noting that the removal rates given in the various studies are not always given in the same unit. Some studies express their findings as the removal of the total load of a HOC in the SS, but most of them express it as the reduction in concentration in the DM. Since a large fraction of the DM is transformed to biogas during the AD, these two removal rates have very different values. Each study's results have been converted into a removal rate expressed as the reduction in concentration on DM basis, given a destruction of 56% of VS and a VS content of 72%. This was also the most common way the studies expressed their findings, so only for a few studies their findings had to be converted. For Triclosan, this resulted in an increased concentration because it was biodegraded at a lower rate than the solids. Lastly, it should also be noted that in some cases the HOCs might degrade to even more toxic compounds, such as the phthalate DEHP which has been confirmed to convert partly into the even more hazardous MEHP [35]. However, due to lack of data and limited timeframe, this is not included in this study. The assumed removal rates are given in table 2.5 below.

Table 2.5: Pollutant removal rates during mesophilic AD.

Pollutant	Removal (% reduction in concentration)	Reference(s)
NP + NPE	12.5	[35–37]
DEHP	27.5	[35, 38]
BPA	65.0	[39]
BDE-209	41.0	[40, 41]
PAH	42.0	[42]
Triclosan	8.0 increase	[43]
PFOS	0.0	[44]
PFOA	0.0	[44]
SCCP	0.0	None found
HHCB	65.0	[45, 46]
AHTN	65.0	[45, 46]
OTNE	65.0	None found, assumed same as the other musks
PCB7	30.0	[47]
LAS	0.0	[48]

HMs, on the other hand, do not biodegrade or volatilize during AD. Consequently, their total load remain constant and their concentration increase in accordance to the solids destruction.

In Norway it is required that SS is sanitized (*hygienisert* in Norwegian) if it is to be used after treatment [26]. The treatment methods that are considered sanitizing steps are highlighted in figure 1.3. Furthermore, for SS to be used in fertilizer products, the temperature should be minimum 55°C during the sanitizing step. Subsequent drying after AD and dewatering should therefore be done if the SS is to be used in fertilizer products. As mentioned, the conventional SS treatment methods are not designed to remove pollutants such as HOCs or HMs, but rather to remove large debris, floating matter, suspended solids, biodegradable organic matter or nutrient salts. Therefore, despite the strict regulations and requirements for SS recycling, there are still concerns for its toxicity and whether it should be allowed to be recycled to soil at all.

2.3.2 Dewatering

Standard for any SS treatment pathway is the use of dewatering. Considering the initial water content of around 95%, dewatering can help to reduce this in an energy efficient way. However, only a DM content of maximum 25-30% is possible to achieve with this method. It is common to also use flocculation agents to help the dewatering process, and according to Cao and Pawlowski [32], dewatering 1 ton of digested SS to a MC content of 73% requires 1.5 kWh electricity, 1.25 kg $FeCl_3$ and 0.263 kg polymer. The two latter were assumed to be "Iron (III) chloride, without water, in 40% solution state GLO| market for | Cut-off, U" and "Polyacrylamide GLO| market for | Cut-off, U", respectively, in the Ecoinvent 3.5 database.

When it comes to the dewatering process, it is assumed that the HOCs and HMs are, in general, strongly sorbed to the SS [20, 49–53], with a few exceptions where a non-negligible share has been found to be removed during dewatering. Consequently, only a very small amount of HOCs is assumed to be lost through the dewatering process. For two of the HOCs (DEHP and BPA), there was available literature on their removal during dewatering. DEHP was found to be removed by 5% from the SS, and BPA by 35% [54, 55]. For most compounds the literature indicated negligible removal. See table C2 in the Appendix for the remaining compounds and the references. No reduction in HM is assumed in this process.

2.3.3 Drying

For this study and all its treatment pathways, drying is necessary to include for two reasons: 1) to sanitize the SS when no pyrolysis is included, and 2) to reduce the water content before pyrolysis, when it is included. Unfortunately, the heat demand is very high for SS drying since the water content needs to be reduced to around 10% of the SS weight prior to pyrolysis.

The energy requirement for SS drying was found using the same method as Cao and Pawlowski [32], because it takes into consideration the MC in the dewatered SS and assumes a MC of 8% in dried SS:

$$Q_{drying} = M_{dw-s} \cdot MC_{dw-s} \cdot \left(Cp_{water} \cdot \Delta T + \Delta H_v\right) + M_{dw-s} \cdot (1 - MC_{dw-s}) \cdot Cp_{SS} \cdot \Delta T \qquad (2)$$

$M_{dw\text{-}s}$ is the flow of dewatered SS into the dryer, Cp_{water} is the heat capacity of water (4.18 kJ/(kg °C)), which is approximated as the heat capacity of the wet SS, ΔT is the temperature difference of the inflowing SS and the drying temperature (10°C and 105°C), ΔH_v is the latent heat of vaporization of water (2257.9 kJ/kg) and Cp_{SS} is the heat capacity of the SS solids (1.95 kJ/(kg °C)) [32, 56]. Finally, a thermal efficiency of 83% was assumed to find the total heat required. Equation 2 gives a total energy requirement of 2.297 MJ per kg SS to the dryer, given the specific process parameters in this study.

No removal nor increase from precursor release is assumed during drying due to little available data and varying drying methods in use. Usually the drying methods use a temperature of around 100°C, so therefore it is valid to assume no destruction or volatilization of HOCs and HMs, due to the high boiling point of both.

To allow for a comparison of soil emissions of HMs based on their bioavailability, the bioavailability of HMs in stabilized and sanitized SS was estimated using the findings in Lu et al. [27]. The bioavailable fractions of HMs were estimated by assuming the same ratio between the actual HM concentration and the bioavailable HM content as found in Lu et al. This ratio might of course not be constant for any concentration, so in reality their bioavailable content could be different. The specific paper was chosen because it estimated the bioavailability both in the treated SS as well as biochar produced from pyrolysis at different temperatures, which made it possible to stay source-consistent and avoid variations in SS characteristics etc. So although the pre-treatment of the SS in this paper (anaerobic-anoxic-aerobic, followed by dewatering and drying) differs from here (anaerobic followed by dewatering and drying), it was assumed that the final bioavailability would be similar. Hg was not included in this study, so the bioavailability of Hg is taken from Janowska et al. [57] where the SS was treated mechanically, biologically (trickling filter) and then dried.

Table 2.6: Assumed bioavailable fractions of the selected HMs in sanitized SS.

Heavy metal	Bioavailable fraction after drying [27]
Pb	1.99%
Zn	14.29%
Ni	5.97%
Cd	4.88%
Cu	5.22%
Cr	4.23%
Hg	0.40%

2.3.4 Pyrolysis

Pyrolysis is a thermochemical process that can transform organic substances to gases, liquids (bio-oil and reaction water) and a solid residue (biochar). It is therefore an option to extract all the remaining energy in the SS into potentially valuable products [58]. Temperatures between 300-1000°C are often used, and the product yields and their composition will vary considerably depending on the temperature. Furthermore, the residence time and heating rate are also important factors for the products [13, 59].

Based on the pyrolysis temperature, residence time and heating rate, one can divide pyrolysis regimes into slow and fast pyrolysis. Slow pyrolysis has a longer residence time, lower heating rate and often lower temperature as well (300-600°C). Generally, the biochar yield is maximized at lower temperatures and low heating rates [9, 13, 56, 59–61]. On the other hand, a higher temperature can increase the recalcitrant carbon fraction in the biochar, i.e. the carbon that will remain in soil on a long-term scale after application [13]. Medium temperature (around 500°C) seems to be the best choice to reduce the biochar's toxicity, likely because of immobilization of HMs and destruction of many HOCs. Interestingly however, studies show that exceeding 500°C can make the biochar's toxicity increase again, due to an increase of toxic compounds that are generated at higher temperatures [62]. In other words, there is possibly a trade-off between the destruction of some HOCs and the generation of other toxic compounds, which is important to keep in mind when determining the pyrolysis temperature if the biochar is intended to be spread on land.

If the biochar is not intended for soil application but rather to be used as a solid fuel, the stability of the carbon is not relevant for the achievable negative emissions. Although it lacks research for SS biochar specifically, the biochar could potentially be used as a solid fuel and replace coal in carbon-intensive industries such as the cement industry. If it is produced in pyrolysis temperatures optimal for maximizing the biochar yield (i.e. around 300°C), the calorific value of the biochar is close to that of low-grade coal with a calorific value of approx. 17 MJ/kg [63]. Since such low-grade coal is phased out in many countries, it is assumed that the biochar could substitute bituminous coal used in the cement industry in Norway in the LCA. However, due to the higher sulphur content in SS biochar it might be challenging in practice to achieve this substitution [12]. Regardless, it is something that is interesting to look into, should it be possible.

Based on the reviewed literature for pyrolysis of SS, slow pyrolysis at a temperature of 500°C is deemed to be the optimal choice for production of biochar to be used in agriculture. This is because of the stability of the carbon and the destruction of HOCs achieved at this temperature, while still having a biochar yield of 33-54% (depending on SS pre-treatment). With higher temperatures the biochar yield would decrease, while there is uncertainty on which benefits this temperature increase would have on the HOCs, HMs and carbon stability. For some HOCs it is possible this would contribute to a higher destruction, but this could be at the expense of a generation of dioxins and furans (PCDD/Fs), PAHs and PCBs, as well as an increased accumulation of HMs [20]. A meta-analysis by Li et al. [64] also showed that biochar produced at higher pyrolysis temperatures could result in reduced crop yields if applied to agricultural land. While it is likely that a higher temperature would give more stable carbon in the biochar, the lower biochar yield means that the negative emission potential would still be lower.

Table 2.7 and 2.8 show the assumed product yields during pyrolysis of both undigested and digested sludge, respectively, at the two chosen temperatures. It was attempted to find references with a similar SS composition as used in this study, since the ash, VS and carbon content are important in determining the product yields and their composition. For the biochar in particular, these are key parameters for the negative emission potential. Due to the complexity of SS as a feedstock, simulation softwares like Aspen Plus could not be used to find yields for the exact composition as chosen for this study. Since other studies have been used instead, with slightly different SS feedstocks, there is not necessarily a carbon balance through the pyrolysis process. The two studies were chosen because they had fairly similar SS compositions, had the same pre-treatments as used in this study, and lastly because they had results for slow pyrolysis at both 300°C and 500°C. As one can see from the yields, the digested SS has a higher biochar yield as % of DM, but since a large part of the DM has already been transformed to biogas, the amount of biochar produced per kg of SS into the system will still be lower. The biochar produced from digested SS also has a lower carbon content than that from undigested SS. Undigested sludge has a much higher yield of reaction water than digested sludge, which could be explained by it being released from decomposition of protein, lignocellulose or fatty acids [60]. Digested sludge has less of these components because they were partly transformed to biogas during AD.

Table 2.7: Yields for slow pyrolysis of undigested SS at the chosen temperatures.

Products	Yields (% DM wt.) for 300°C	Yields (% DM wt.) for 500°C
Biochar	52	33
Bio-oil (dry)	10	18
Syngas	3	4
Water	35	45

[9]

Table 2.8: Yields for slow pyrolysis of digested SS at the chosen temperatures.

Products	Yields (% DM wt.) for 300°C	Yields (% DM wt.) for 500°C
Biochar	75	54
Bio-oil (dry)	14	26
Syngas	4	8
Water	7	12

[56]

The compositions and calorific values of the bio-oils and syngas can be found in table A1 and A2 in the Appendix. The composition of the biochar produced from non-digested SS was taken from the same study as the pyrolysis yields, since they had a similar SS composition and it was the only study using non-digested SS pyrolysis and the specific pyrolysis conditions [56]. The study used for the yields of slow pyrolysis of digested SS did not include the biochar compositions, and therefore other references with other SS compositions were used to determine its carbon content and caloric value (the key parameters for negative emissions and coal substitution potential). Jaramillo-Arango et al. [60] had a lower carbon content than the digested SS in this study, while Tang et al. [65] had a higher carbon content than here. The average of the carbon contents in the biochars produced under slow pyrolysis at 500°C in these two studies is therefore assumed for the biochar produced in this study. Since neither of these papers

Table 2.9: Key parameters for the biochar produced from digested and non-digested sludge under the chosen pyrolysis conditions.

	Undigested SS		Digested SS	
Biochar properties	300°C	500°C	300°C	500°C
C content	45.4	40.5	18.0	14.5
HHV (MJ/kg)	18.6	16.0	15	n/s

included the calorific value of the biochar, this was taken from Pulka et al. [12] where they plotted the HHV of biochars based on the pyrolysis temperature. In other words, it does not take into account the potential pre-treatment and might therefore be inaccurate for undigested sludge. Below, in table 2.9, is a summary of the key parameters for the biochars relevant for this study.

Slow pyrolysis with a long retention time seems to be of importance for the removal of some of the HOCs. For example, the sum of estrogenic substances including, among many others, BPA, NP+NPE and Triclosan, was only significantly reduced in a study by Hoffmann, T. after 60 minutes [66]. In another study however, Ross, J. et al. found that after only 5 minutes NP+NPE and Triclosan were below detection limit. The study by Hoffman, T. was the only available study including the effect of SS pyrolysis on BPA, so therefore it was chosen as reference for BPA even though it only measured the sum of estrogenic substances. It is possible that BPA would degrade faster if measured individually in this study. PFOS and PFOA was kept at peak temperature of 700°C for 3 hours in a study by Kim et al. [67], and yet it was not found to be destroyed at a higher rate than the feedstock volatilization, thus the concentration in the biochar was the same as in the dried SS.

Unfortunately, the studies used for HOC degradation during pyrolysis are quite inconsistent in how they state the pyrolysis conditions. Some studies specify the heating rate, others specify the retention/residence time, and others again specify both heating rate as well as an additional residence time at peak temperature. Table 2.10 therefore includes the heating rate and the residence time, whichever is specified, for the chosen HOCs and their removal during 500°C pyrolysis. As evident from the table, most of the compounds are either completely destroyed or their concentration remains unchanged compared to the SS concentration. The exception is LAS, which was found to decrease in mass by 42% after pyrolysis. For LAS, the literature was very scarce and the only study found did not study the fate of LAS during SS pyrolysis, so its actual destruction in SS pyrolysis remains uncertain. The same goes for SCCP and BDE-209; since they are not in SS in the reference studies found, their destruction might be different when sorbed onto SS solids.

The effect of pyrolysis at 300°C and 500°C for the HMs will, as mentioned, be estimated by using the effect of pyrolysis on their bioavailability and the retention rates (fraction of HMs in SS feedstock ending up in the biochar) from Lu et al. [27]. The bioavailable fraction of each HM will be multiplied by the estimated mass of the respective HM to find the resulting bioavailable soil emissions for each treatment scenario. Their results indicate that pyrolysis would reduce the bioavailability of the HMs to around half for Pb, Zn and Ni, and even more for the others. The bioavailable fraction of each HM after SS pyrolysis at the chosen temperatures is given in table 2.11. Hg is a metal with a very low boiling temperature and will therefore volatilize at around 300-400°C. No studies with its retention rate or bioavailability in biochar from 300°C was found, and because of its low concentration and bioavailability in SS (0.40%), its retention rate is assumed to be 0% during pyrolysis.

The energy consumption of pyrolysis was estimated using the method from Kim and Parker [56], which takes into account (1) energy requirement to raise the incoming SS to the final pyrolysis temperature, (2) reaction heat of pyrolysis, and (3) heat loss through pyrolysis reactor. The heat loss was assumed to be 10% of the total energy consumption, and the reaction heat, $Q_{pyrolysis}$, was assumed to be 300 kJ/kg.

Table 2.10: Assumed HOC removal rates as % reduction in biochar concentration from the initial concentration in the feedstock, during pyrolysis at 500°C. Residence time and heating rate where it is specified in the study.

HOC	Removal (%)	Heating rate (°C/min)	Total residence time (minutes)	Reference(s)
NP + NPE	99	-	5	[68]
DEHP	0	-	-	[69]
BPA	100	-	60	[66]
BDE-209	100[a]	10	-	[70]
PAH	99	-	400	[71]
Triclosan	100	-	5	[68]
PFOS	0	7	400	[67]
PFOA	0	7	400	[67]
SCCP	100[a]	10	-	[72]
HHCB	99[b]	-	-	[20]
AHTN	99[b]	-	-	[20]
OTNE	99[b]	-	-	[20]
PCB7	97		30	[73]
LAS	42[a,c]	-	180	[74]

[a]The rate is based on a study with a different feedstock. [b]Assumed in reference study. [c]On total mass basis, meaning a potential concentration increase in the biochar depending on the yield.

Table 2.11: Assumed bioavailable fractions of the selected HMs in SS biochar.

	Pyrolysis 300°C		Pyrolysis 500°C	
HM	Retention rate	Bioavailable fraction	Retention rate	Bioavailable fraction
Pb	98.3%	1.08%	95.1%	0.97%
Zn	99.2%	9.62%	98.3%	6.77%
Ni	99.3%	2.56%	93.8%	2.10%
Cd	97.1%	1.6%	96.6%	1.23%
Cu	94.5%	0.15%	86.9%	0.10%
Cr	87.5%	1.59%	80.6%	1.27%
Hg	0.0%	n/a	0.0%	n/a

The heat requirement for feedstock heating was calculated using the following equation

$$Q_{target} = M_{dr-s} \cdot Cp_{SS} \cdot \left(T_{final} - T_{dr-s}\right) \tag{3}$$

As mentioned, the yields and composition of the pyrolysis products are highly dependent on both SS characteristic and process conditions. The specific product yields for the two systems are given in table 2.7. The composition of syngas is also highly temperature and feedstock dependent, but due to lack of data it was assumed to be the same for undigested and digested SS. Its compositions at the two temperatures are in table A2 in Appendix A. The retention time in the pyrolysis reactor was varying in the different studies used as references, and in some cases not specified. This can lead to inaccuracies in the results, since the products and their compositions depend on both the temperature and the pyrolysis time. Only data from references using slow pyrolysis at 300°C and 500°C have been used, however the exact heating rates and and retention times are not the same in each of the studies.

2.3.5 Soil application of SS biosolids and biochar

When SS biosolids (i.e. treated and dried SS) and biochar are applied to land, they can have emissions to air, soil and water, depending on the application site. As a simplifying assumption, it is assumed that the emissions of HMs and HOCs are only to soil due to the uncertainty of how large fraction of them would end up in groundwater and/or rivers. It is also assumed the HOCs will not volatilize and be emitted to air. When biosolids are applied to land, they will also decompose and emit GHGs [6], both directly and indirectly. The GHG emissions hold a significant uncertainty because they depend on the SS composition and treatment, as well as site-specific conditions such as application method, soil type and the local climate.

A transport distance of 150 km by road (round-trip) from the treatment plant to the application site of biosolids and biochar is assumed. Since there was no explicit data or literature to use for this estimate, 150 km is assumed to reflect the restrictions on land application of SS-derived products and the limited coal substitution possibilities in Norwegian industries. The transport process is taken from the Ecoinvent 3.5 database and is the process "Transport, freight, lorry 16-32 metric ton, euro5 RER| market for transport, freight, lorry 16-32 metric ton, EURO5 | Cut-off, U".

The process "Solid manure loading and spreading, by hydraulic loader and spreader GLO| market for | Cut-off, U" in Ecoinvent 3.5 is used to model the spreading of the biosolids and biochar to soil. This process does not include any emissions from the manure itself, but rather the emissions related to diesel fuel consumption, agricultural machinery and emissions from combustion and tyre abrasion, per kg manure spread.

Soil application of SS biosolids

To estimate the GHG emissions of biosolids application to land, IPCC Good Practice Guidance for national GHG inventories [6] recommends to use country-specific emissions if the country has rigorously documented country-specific values for estimating these emissions. Since this does not exist for Norway, the default IPCC values are used to calculate the direct and indirect emissions of N_2O from biosolids application to land. While SS land application also emits small amounts of methane and NMVOCs, they are excluded in this method since they are relatively unsignificant. This is also in accordance with the methodology of the Norwegian emission inventory [7]. In the national emission inventory, IPCC Tier 1 methodologies and default emission factors are used with the N-content of treated SS as reported by Statistics Norway (SSB) to estimate emissions the N_2O from SS application to agricultural soil. As previously shown in figure 1.4 in the Introduction, this is the most common disposal method of SS in Norway.

The total N_2O emissions from SS application to agricultural land can be expressed generally as

$$N_2O_{SS} = N_2O_{direct} + N_2O_{indirect} = N_2O_{direct} + N_2O_{(G)} + N_2O_{(L)} \tag{4}$$

where N_2O_{direct} represents the direct N_2O emissions as a result of SS application to soil, and $N_2O_{indirect}$ is the sum of the indirect N_2O emissions produced from volatilization of the SS N and its subsequent atmospheric deposition as NO_x and NH_3, $N_2O_{(G)}$, plus the N_2O produced from leaching and runoff, $N_2O_{(L)}$.

Applying the Tier 1a methodology, the direct N_2O emissions from SS application to agricultural soils can be estimated as follows:

$$N_2O_{direct} = N_{SS} \cdot EF_1 \tag{5}$$

where N_{SS} is the amount of SS nitrogen applied to soil (kg N input) and EF_1 is the emission factor for emissions from N inputs (kg N_2O-N/kg N input). See Appendix E for the values.

Continuing using the Tier 1 methodology, here using the equation in Tier 1b, the indirect N_2O emissions from volatilization of the SS N and its subsequent atmospheric deposition as NO_x and NH_3 can be found using the following equation:

$$N_2O_{(G)} = N_{SS} \cdot Frac_{GASM} \cdot EF_4 \tag{6}$$

where N_{SS} is the total amount of N (kg) deposited by SS application to land, $Frac_{GASM}$ is the fraction of SS N that volatilizes as NH_3 and NH_3 (kg NH_3-N and kg NO_x-N/kg of N input) and EF_4 is the emission factor for atmospheric deposition (kg N_2O-N/kg NH_3- and NO_x-N emitted).

Lastly, the Tier 1 methodology for estimating the N_2O produced from leaching and runoff is as follows:

$$N_2O_{(L)} = N_{SS} \cdot Frac_{LEACH} \cdot EF_5 \tag{7}$$

where N_{SS} is the total amount of N (kg) deposited by SS application to land, $Frac_{LEACH}$ is the fraction of N input that is lost through leaching and runoff and EF_5 is the emission factor for leaching/runoff (kg N_2O-N/kg N leached/runoff).

These emissions should be taken with caution because of the complexity of these emission pathways. In particular the default IPCC factors, as they have not been updated since 2006 and more recent studies indicate that they might underestimate emissions [75, 76]. The methodology with its default factors is used worldwide regardless of variations in the applied SS and its pre-treatment, soil type, climate, etc. For the temperate climate in Norway it is possible they overestimate emissions. Initially, it was attempted to use some Danish factors as estimated through an experimental study of applying different kinds of treated SS to Danish agricultural land [77]. Using their factors gave lower N_2O emissions than using the IPCC default factors. However, this method was discarded since the IPCC only recommends country-specific values if they are rigorously documented for the specific country, which is not the case for Norway. A sensitivity analysis for this is included since emissions of N_2O have a GWP100 of 265 times to that of CO_2 [2], so even small changes in its emissions can have large impacts on the GWP category.

The same Ecoinvent processes for transport and spreading of biosolids to land is assumed for biochar.

Some LCA studies also include a substitution of synthetic fertilizers when applying SS to agricultural land. This could have been implemented here as well, but was excluded due to lack of time in finding a good methodology in line with the Norwegian SS regulations restricting its use on farmland.

Soil application of SS biochar

The main benefit of biochar application to soils is the carbon sequestration potential, but there could be several co-benefits depending on site-specific conditions. In contrast to application of SS biosolids, the application of SS biochar could possibly be able to *reduce* soil GHG emissions while at the same time improving soil quality and yields. While there has been extensive research on the effect on soil emissions of biochar application from other feedstocks, there is limited research on the effect of SS biochar. That being said, numerous studies have found that SS biochar can improve soil quality and crop/plant yields in various soil types [78–84].

Starting with the biochar's negative emission potential, we can estimate this from the fraction of its carbon that can be considered stable after soil application. It can be assumed from proximate analyses that approximately 20% of the SS biochar's carbon is volatile, meaning that it is quickly degraded after application to soil [13, 16]. The remaining fraction, 80% of the carbon in the biochar, is recalcitrant and can remain in the soil on a centennial scale. Using this assumption along with the carbon content per kg biochar, as shown in table 2.9, one can calculate the potential negative emissions.

The impact SS biochar can have on soil GHG emissions, soil quality and crop yields is strongly site-specific. There is unfortunately little available research on SS biochar application in or near Norway, as the literature is mostly concentrated in tropical areas with very different conditions than in Norway. Considering that experiments with other biochar feedstocks show highly varying impacts of its land application depending on application area, it would not be reasonable to use findings from experiments in areas like Brazil or Australia to estimate the effects in Norway [17].

A biochar feedstock that has been more researched in terms of its effect on soil GHG emissions and crop yields is manure. Liu et al. [17] used Random Forest regression modelling of machine learning and data from the literature to map the spatial variability of the impacts of different biochar types and their application rates. The impacts included the same soil N losses as the methodology used by the IPCC for biosolids: NH_3 volatilization, N_2O emissions and N leaching.

While it is not a perfect assumption, the best method to estimate the effects of SS biochar application to Norwegian soil (other than the carbon sequestration) was deemed to be using manure biochar as a proxy for SS biochar. Although they have different physicochemical properties, their effects on the soil seem to be quite similar. Li et al. [64] used data syntheses to link biochar feedstock and pyrolysis temperatures to their effect on N retention, crop yield and N_2O emissions, and the results indicate that manure and SS biochar have very similar effects, with SS biochar performing slightly better. It therefore seems like a reasonable assumption that will not give an overestimation, but possibly rather an underestimation, on the co-benefits of SS biochar application to land.

Table 2.12: Findings from Liu et al. used to estimate SS biochar effect on soil.

Category	Current emissions (kg N/ha)	Value used in LCA (kg N/ha)
NH3 volatilization	10-15	12.5
N2O emissions	3-4	3.5
N leaching	25-30	27.5
Category	Range of change	Value used in LCA (average)
Crop productivity	+ 4-12%	+ 8%
NH3 volatilization	+ 40-50%	+ 45%
N2O emissions	- 12-24%	- 18%
N leaching	- 0-4%	- 2%

By zooming in over Norway on the maps made by Liu et al. [17], one can retrieve their findings on the effect of manure biochar application in Norway on crop productivity, soil NH3 volatilization, soil N2O emissions and soil N leaching. Because of the strict regulations on application of SS derived products to agricultural land in Norway (very low application rates are allowed per hectare), the lowest application rate in Liu et al. of 0-10t/ha was used. It also made sense to choose this because, based on their data syntheses, the authors recommend very low application rates of manure in and near Norway to optimize the effect on soil N losses and crop yield. A higher application rate of manure biochar in Norway results in such a high increase in NH_3 volatilization that the net effect on N_2O emissions might be an increase. It also seems like the application rate of 0-10tons/ha gives approximately the same increase in crop productivity as 10-20tons/ha, further motivating this choice of application rate. Table 2.12 summarizes the data taken from Liu et al. for estimating the change in N_2O emissions given the chosen application rate and feedstock proxy.

Similarly as for the N_2O emissions from SS biosolids application, the N_2O emission reduction from SS biochar application can be found using the following method:

$$N_2O_{net\,effect} = -N_2O_{red.\,(direct)} - N_2O_{red.\,(L)} + N_2O_{inc.\,(G))} \tag{8}$$

where the reduction in direct N_2O emissions, $N_2O_{\mathrm{red.\ (direct)}}$, is the % change as defined in table 2.12 multiplied by the current N_2O emissions, and the reduced emissions from N leaching can be found using the default IPCC factors again:

$$N_2O_{red.\,(L)} = N_{red.(L)} \cdot EF_5 \tag{9}$$

where $N_{\mathrm{red\ (L)}}$ is the reduced leaching (kg N) found by multiplying the current N leaching with the assumed reduction percentage and EF_5 is the emission factor for leaching/runoff (kg N_2O-N/kg N leached/runoff).

The increased N_2O emissions as a result of the increased NH_3 volatilization can be calculated using the same approach as above and the default emission factor for NH_3 volatilization:

$$N_2O_{inc.\,(G)} = N_{inc.(G)} \cdot EF_4 \tag{10}$$

where $N_{inc.\ (G)}$ is the increased NH_3 volatilization (kg NH_3) found by multiplying the current NH_3 with the assumed reduction percentage and EF_4 is the emission factor for atmospheric deposition (kg N_2O-N/kg NH_3- and NO_x-N emitted).

As for the fertilizer substitution potential of biochar, several assumptions had to be made. From the maps in Liu et al. [17] it seemed that the effect on crop productivity in Norway was similar for application rates between 0-10 tons/ha manure biochar and for 10-20 tons/ha. Based on this, and the findings in Li et al. [64] suggesting that 1) the effect on crop productivity is higher for biosolids than manure, and 2) the effect seems to be maximized at pyrolysis temperatures between 401-500°C, it seems reasonable and conservative to use the assumption from Gaunt and Lehmann [85] that the fertilizer can be reduced by 10% with the addition of biochar. The fertilizer application rates were taken from Yara's suggested application rates for grains in Norway (one of the few crops that SS-derived fertilizers can be applied on) [86] and their conversion calculator to kg N, P_2O_5 and K_2O. Next, the reduced N_2O emissions as a result of reduced N fertilizer application was calculated on the basis of the IPCC default methodology for synthetic N fertilizers [6] as done in Cherubini and Jungmeier [87], where they found that 1.325% of N in synthetic fertilizers is emitted as N in N_2O. See Appendix E for more detail on the calculation.

2.4 Use of biogas and pyrolysis products for heat, electricity and fuel substitution

2.4.1 Use in combined heat and power (CHP) system

The easiest way to take advantage of the energy content in the biogas, bio-oil and syngas is to use it in a CHP system. This is because the use in a CHP does not require any upgrading of the products, and since the treatment systems themselves have a high heat and electricity demand, the majority can be used internally, and any excess electricity can be exported to the grid (the excess heat could have been exported to an adjacent user, but there is never any excess heat).

The possible heat and electricity production from the various products were estimated based on their calorific values as shown in table 2.14 and the electrical and thermal efficiencies of the CHP system, taken from the Patterson et al. [88]. The emissions to air from the operation of the CHP system is taken from Li et al. [21], and to simplify the calculations, they are assumed to be the same per kWh generated, regardless of products used in the CHP. The efficiencies and emissions are given in table 2.13.

Table 2.13: CHP conversion efficiencies and emissions used in this study.

CHP conversion efficiencies and emissions	
Electrical	32 %
Thermal (heat)	50 %
CO (mg/kWh)	986
NOx (mg/kWh)	821
SO2 (mg/kWh)	439
NMVOC (mg/kWh)	136

2.4.2 Biogas upgrading to biomethane

An alternative use of the biogas is to upgrade it so that it is nearly 100% pure methane, and therefore can be used as a substitute to natural gas (NG). Upgrading of biogas to biomethane requires electricity

Table 2.14: Calorific values for the products in this study.

Product	Treatment pathway	LHV (MJ/kg)	Reference
Biogas	-	25.8	[31]
Bio-oil (dry)	AD+PY500	35.2	[89]
Bio-oil (dry)	AD+PY300	33.2	[89]
Bio-oil (dry)	PY500	34.5	[9]
Bio-oil (dry)	PY300	24	[9]
Syngas	AD+PY500	9.119	[60][a]
Syngas	AD+PY300	0	[60]
Syngas	PY500	9.119	[60][a]
Syngas	PY300	0	[60]
Biochar	AD+PY500	-	
Biochar	AD+PY300	15[b]	[12]
Biochar	PY500	-	
Biochar	PY300	18.6[b]	[9]

[a] Using net calorific values and densities from [90] for the individual gas components.

[b] Values are HHVs.

for the removal of the CO_2, and often includes a small loss of the methane in the process. The separated CO_2 is assumed to be released to the atmosphere in these scenarios, but since it is of biogenic origin it is not considered to increase the GWP [2]. From an industrial ecology point of view, the optimal scenario allows for reuse of the CO_2, but this will not be considered in this study. One widely used biogas upgrading technology is high pressure water scrubbing [91]. This technology is assumed to be used in the scenarios in this study, and table 2.15 summarizes the parameters that will be used for the inventory, all of which are taken from Starr et al. [91]. The upgraded biogas is assumed to substitute NG on an energy equivalence basis where the NG is considered to have an LHV of 37 MJ/Sm^3 [92] and the biomethane has an LHV of 49 MJ/kg.

Table 2.15: Data used for biogas upgrading with high pressure water scrubbing.

Biogas upgrading	
Electricity (kWh) per 1000 kg CO_2 removed	213,89
Methane purity (%)	98
Final LHV (MJ/kg)	49
Methane loss in upgrading (%)	1.0

[91]

After upgrading, the biomethane can then be injected into the gas grid. Due to uncertainties in its final use, the LCA will only consider the fossil CO_2 emissions as avoided burdens, along with the natural gas it can substitute on an energy basis. More about this is described in section 2.5.1.

2.4.3 Bio-oil upgrading to fuel quality

Upgrading bio-oil from SS to diesel or gasoline quality is an option that has little experience as of today, for several reasons. One reason is that SS pyrolysis is not a widely used technique, so the possible application of the bio-oil has not been studied as extensively as for other feedstocks. Moreover, SS bio-oil has some undesirable fuel properties such as high fractions of oxygenated and nitrogenated compounds, high viscosity and high instability [93]. Even so, due to the need for more sustainable liquid fuels to mitigate climate change, it is of interest to include scenarios with bio-oil upgrading to assess its impacts, albeit if it remains hypothetical.

One method that can be used to upgrade the bio-oil is by using H_2 to hydrotreat and hydrocrack the oil [94]. In simple terms, the hydrotreatment can produce a stable hydrocarbon oil with a low fraction of oxygenated compounds, a process requiring some H_2 under pressure and at elevated temperatures. Next, the heavy hydrocarbon fraction from the hydrotreated oil is cracked to additional gasoline and diesel blendstocks [94, 95]. The LCA will not be able to capture the complexity of this upgrading process completely, but a simplified inventory of the necessary inputs to the system will be estimated.

The key inputs to the upgrading process are H_2 and electricity. While there is already H_2 in the syngas produced at 500°C pyrolysis, the fraction of the H_2 in the syngas is uncertain and might vary considerably depending on feedstock and process conditions, and at lower temperatures the syngas is almost entirely CO_2. Therefore, although theoretically possible in the pathway with 500°C pyrolysis, the possibility of separating the H_2 from the syngas is not considered. Instead, the commonly applied method of using steam methane reforming (SMR) to separate H_2 from NG is assumed in this study. The electricity for the H_2 compression is estimated based on the plant-specific conditions in Da Silva et al. [96]. The higher flowrate of bio-oil through the upgrading system, the lower electricity consumption per kg H_2 compression, so in the case of a lower bio-oil production, the electricity consumption might be much higher. Due to the different yields of bio-oil in the various systems this would complicate the calculations to include, so a constant electricity consumption per kg H_2 is assumed. See Annex F for the calculation of the specific electricity consumption, and see table 2.16 for the parameters used to estimate electricity and NG demand.

Table 2.16: Parameters for steam methane reforming and bio-oil upgrading used in this study.

Parameter	Value	Unit	Reference
NG cons.	4,8 %	kg NG/kg bio-oil	[95]
Electricity for H_2 prod.	67	kWh/ton H_2	[96]
Electricity for H_2 comp.	2062	kWh/ton H_2	Estimated from [96]
Rate of H_2 in upgrading	5.80%	kg H2/kg bio-oil (dry)	[94]
Emissions of CO_2	7.0	kg CO_2/kg H_2	[97]

2.4.4 Biochar used as coal substitute

The use of biochar as a coal substitute is also more of a hypothetical option, as it lacks testing and research. This study will only explore the LCA impacts if it were to be feasible, for the purpose of looking into other disposal methods that can also contribute to abatement of the hazardous compounds, as well as negative emissions. Since the temperatures during coal combustion are so high, it is likely that most of, if not all, the HOCs will be destroyed.

It is here assumed that the biochar produced from pyrolysis at 300°C can be used directly to substitute coal in a cement kiln in Norway. Moreover, it is assumed that the cement kiln has implemented oxy-fuel combustion to allow for CCS and sequestration of impurities [98]. The CO_2 released during combustion of biochar is assumed to be 100% biogenic [2].

After combustion of the biochar, what will remain is likely just the fly- and bottom ash where most of the HMs are still present [98]. The exact fate of this ash might vary between industries, but here it is assumed that it is spread on land, as often done with the ash from SS incineration [99]. Due to the uncertainty of the fate of the ash, as well as uncertainty of its final composition, only a simplified estimate of the soil emissions of HMs from land spreading is included. Furthermore, it is assumed that the bioavailability of the HMs are the same in the fly- and bottom ash as in the original biochar because of lack of data suggesting otherwise. A study by Tang et al. [100] also indicates low ecological risk and low leachability from the fly ash from co-combustion of SS (note: not biochar) and coal. It is assumed that the high temperature in the coal combustion plant would be sufficient to destroy the HOCs in the biochar, and no emissions of HOCs are therefore considered from the ash.

The electricity for operating the oxy-fuel combustion plant is not included since it is assumed that it is already implemented in the cement kiln. That being said, the electricity is usually on a per-kg-CO_2-captured basis, and since the amount of carbon per kg (and per MJ) of biochar is much lower than for coal, the process would likely require less electricity input using biochar as a substitute [101]. Table 2.17 shows the parameters used for estimating the inventory for biochar substitution for coal.

Table 2.17: Parameters for coal substitution and oxy-fuel combustion for CCS assumed for this study.

Parameter	Unit	Value	Reference
C capture efficiency	% C	96	[102]
HHV of bituminous coal	MJ/kg	25	
C content of biochar	kg C/kg biochar	45.4-14.5	[12, 56, 60, 65]
kg C to kg CO2	-	44/12	Molecular weights

2.5 Life cycle assessment

Life cycle assessment (LCA) is a science-based tool that has been developed for assessing the environmental impact of products and services over their life cycle.

The standardized method includes four phases: 1) goal and scope definition, 2) life cycle inventory (LCI) analysis, 3) life cycle impact assessment (LCIA) and 4) interpretation [103]. This holistic approach allows for quantification of the environmental performance of products and services in an objective manner.

2.5.1 Goal and scope definition

As described in section 2.1, the goal of the study is to compare the environmental impacts of different SS treatment scenarios for implementation in Norway with a reference case for Norwegian SS management. The scenarios are described in table 2.1 and figures 2.1 and 2.2.

The function of the system should be captured by the functional unit (FU) of the study, an important element of an LCA providing the basis on which the inputs and outputs are related and compared. The FU for this study is chosen as treatment of 1 ton DM SS per hour (i.e. 20 tons raw SS given the MC). This unit is chosen to remain in line with the function of a SS treatment system, i.e. treatment of a specific amount of SS with a given capacity.

The system boundaries for the systems are illustrated in figure 2.2. Each system includes (1) the entire SS treatment from it enters as raw SS, (2) transport of biosolids or biochar to use site (3) end use of biogas, bio-oil and syngas including the necessary upgrade, and (4) biochar management. Construction and dismantling of the treatment plants are excluded from this LCA because they generally have large uncertainties, and other studies find their impacts to be negligible in comparison to those in the operation period [21, 32, 104].

As for the products with potential to be used for production of heat or electricity or replace other existing products and services, a system expansion is done where they are considered as alternatives to the commonly used product. That way, they are regarded as avoided burdens in terms of how much of a product they can replace. This is also shown in figures 2.1, 2.2a and 2.2b, where the system includes the benefits of the co-products credited from substitution of other fossil fuels, heat and electricity from from fossil fuels or substitution of synthetic fertilizer.

2.5.2 Inventory analysis

In the inventory analysis (LCI), all the data related to the system and its FU is collected and quantified. When the system has a multi-functionality, i.e. the system products and co-products can substitute products that already exist in the market with a function, this is also quantified and assessed as avoided products to the technosphere. As mentioned, SimaPro and the library "Ecoinvent 3.5 - allocation, cut-off by classification - unit" is used for the inventory products and processes. The complete inventory per FU in each scenario is shown in table F1 in the Appendix.

Substituted natural gas
In the scenario where biogas is upgraded to biomethane, it is assumed to substitute the product "Natural gas, high pressure NO| market for natural gas, high pressure | Cut-off, U" in the Evoinvent database. The substitution was calculated based on the assumed LHV of 50 MJ/kg in methane and a 98% (weight basis) of methane in the biomethane, and 37 MJ/Sm^3 for Norwegian NG [92]. Furthermore, based on the emission intensity of NG of 56.1 g CO_2 per MJ, the fossil emissions of fossil CO_2 were subtracted [105].

Substituted diesel
Similarly, the diesel substitution offered by upgraded bio-oil is calculated based on the net calorific values (LHVs) of the bio-oil, as shown in table A1, and substitutes "Diesel Europe without Switzerland| market for | Cut-off, U" assuming a LHV of 45.50 MJ/kg diesel [106]. Due to uncertainty in the emissions of potentially toxic compounds from combustion of upgraded bio-oil, it was assumed to be similar as for diesel, and only the fossil CO_2 emissions were considered as avoided emissions. Some contaminants from the SS end up in the bio-oil during pyrolysis and may escape during its combustion [22]. The emission factor for diesel from IPCC, 0.0741 kg CO_2/kg MJ diesel burned, was used to calculate the avoided fossil CO_2 [105]. A more careful assessment of this substitution, considering emissions of particulate matter etc., is thus possible to do in future work.

Substituted coal
The substitution of coal was calculated assuming that the biochar can replace bituminous coal on an energy equivalence basis. The references used for the biochar composition and properties only give the HHVs of the biochar. Since biochar seems to have a similar content of moisture and hydrogen as bituminous coal [9, 60, 65, 107], the HHVs of the biochars and bituminous coal are used to estimate how many kg of coal can be replaced by the produced biochar. The most similar product in Ecoinvent is "Hard coal Europe, without Russia and Turkey| market for hard coal | Cut-off, U" with an assumed HHV of 25 MJ/kg [108] (varies between types of hard coal as well). Since it is assumed that CCS and sequestration of impurities were already implemented on the combustion site, there were no subtracted CO_2 emissions or any other emissions from coal combustion here - only the negative emissions from biochar.

Substituted electricity/heat
The substituted heat and electricity calculated using the LHVs of the products and the CHP conversion efficiencies are modelled as avoided "Heat, central or small-scale, natural gas Europe without Switzerland| market for heat, central or small-scale, natural gas | Cut-off, U" and "Electricity, high voltage NO| heat and power co-generation, natural gas, conventional power plant, 100MW electrical | Cut-off, U" to the technosphere in SimaPro.

2.5.3 Impact assessment

The impact assessment (LCIA) translates the data from the LCI into environmental impacts in a set of impact categories. Depending on the impact assessment method and its main focus, there can be different impact categories. For this study, the ReCiPe 2016 Midpoint (H) method is used for the majority of the impact categories. This is the hierachist perspective of the ReCiPe methods, and since it is based on scientific consensus regarding the time frame and plausibility of impact mechanisms it is considered the default ReCiPe midpoint method [109]. Moreover, midpoint characterization is chosen because of the relatively low uncertainty compared to endpoint. For the impact categories relevant for HOC emissions, the USEtox 2 method is used instead in order to include those HOCs that had to be calculated outside of SimaPro using USEtox characterization factors and units. Table 2.18 shows the impact categories to be used in the LCA. That allowed for a summation of the results from SimaPro and the results calculated outside SimaPro.

Table 2.18: Impact assessment methods and categories used in this LCA.

Impact categories from the ReCiPe (2016) midpoint (H) method		
Category	Abbreviation	Unit
Global warming	GWP	kg CO2 eq.
Stratospheric ozone depletion	ODP	kg CFC11 eq.
Ionizing radiation	IRP	kBq Co-60 eq.
Ozone formation, Human health	HOFP	kg NOx eq.
Fine particulate matter formation	PMPF	kg PM2.5 eq.
Ozone formation, Terrestrial ecosystems	EOFP	kg NOx eq.
Terrestrial acidification	TAP	kg SO2 eq.
Freshwater eutrophication	FEP	kg P eq.
Marine eutrophication	MEP	kg N eq.
Terrestrial ecotoxicity	TETP	kg 1,4-DCB
Marine ecotoxicity	METP	kg 1,4-DCB
Land use	LOP	m2a crop eq.
Mineral resource scarcity	MDP	kg Cu eq.
Fossil resource scarcity	FDP	kg oil eq.
Water consumption	WCP	m3
Impact categories from the USEtox 2 (recommended + interim) V1.0 method		
Category	Abbreviation	Unit
Human toxicity, cancer	HTPc	cases
Human toxicity, non-cancer	HTPnc	cases
Freshwater ecotoxicity	FETP	PAF.m3.day

Some of the selected HOCs lack characterization factors in both the ReCiPe 2016 Midpoint (H) method and the USEtox 2 method. That leaves two other options: finding suggested characterization factors in the available literature, or, if that is not possible either, attempt to calculate them using the USEtox 2 user inteface wizard and data from the literature. The last option is naturally the method that would give the least accurate characterization factors, due to the limited timeframe of this study and the limited toxicity knowledge of the author.

After searching in the USEtox database in SimaPro, as well as the USEtox software itself, the HOCs that do not have characterization factors in USEtox are: PFOS, PFOA, SCCP, HHCB, AHTN and OTNE. In addition, USEtox operates with individual PAHs and not a group of PAHs, like PAH_{16} used here. The quantification of PAHs through the systems has been done looking at the sum of PAH_{16}, however the resulting PAH emissions will be split into the 16 individual PAHs based on their average initial concentration, taken from a report by Norsk Vann in 2018 [24], assuming that they are destroyed at an equal rate during the treatment processes. This is a simplification and is likely not accurate as they may be biodegraded and destroyed at different rates. The individual PAHs and their assumed initial concentration

Table 2.19: The characterization factors for the toxicity of the substances not found in the USEtox database.

Substance	HTPc (cases/kg)	HTPnc (cases/kg)	FETP (PAF.m3.d/kg)
PFOS	0.0000111	0.00414	592
PFOA	0.0000111	0.00414	592
SCCP			3870
HHCB			41.28
AHTN			1.993
OTNE			14.451

can be found in Appendix B.

For PFOA and SCCP, estimations of their characterization factors were available in the literature. Holmquist et al. [110] calculated the toxicity characterization factors for several PFASs, including PFOA. The same factors are assumed for PFOS since, according to the European Food Safety Authority, *"they share toxicokinetic properties and show similar accumulation and long half-lives. Also, in terms of effects, these compounds in general show the same effects when studied in animals."* [111]. The characterization factor for SCCP is taken from Kobetivcova et al. [112], where they calculated the freshwater ecotoxicity (FETP) factor. See table 2.19 for the factors.

Unfortunately, there were no studies with characterization factors for the three musks included in this study. This may be because, due to their large use in household products and cosmetics, they are believed to pose little to no risk for human health. That being said, both HHCB and AHTN are currently under assessment as endocrine disrupting by the European Chemicals Agency [113]. Their toxicity to other ecosystems is the main concern for these HOCs since they are found at high quantities in wastewater. Therefore, it was attempted to calculate the FETP using the USEtox software, the USEtox manual for organic substances [114] and the data available. USEtox recommends using experimental data whenever it is available, and if not, predicted data in EPIsuite may be used. Experimental data was found for most of the properties of HHCB and AHTN, but not for the degradation rates of OTNE. Initially, it was therefore attempted to use the Biowin 3 conversion factors as described in the USEtox manual [114]. This gave an unusually high FETP of OTNE (as high as PFOA, which is in the top 5% in FETP in the USEtox database). OTNE is found in a much higher concentration than the extremely toxic PFOS and PFOA in SS, it therefore seemed unlikely it is equally toxic. Moreover, since the predicted degradation based on the Biowin 3 conversion factors for AHTN and HHCB was much lower than the experimental values (i.e. much slower degradation), this further motivated the use of different degradation values. The Biowin 3 values indicated a similar degradation time of "months" for all three musks. Therefore, for OTNEs degradation in water, sediments and soil, the averages of the experimental values for HHCB and AHTN were used instead. No studies on the bioaccumulation of OTNE were available either, so the averages of the bioaccumulation factors for HHCB and AHTN were again assumed for OTNE. All the parameters and their references can be seen in table G1 in the Appendix.

2.5.4 Interpretation

The last phase of LCA, interpretation, considers all previous phases in order to interpret the results. To get a clearer picture of the process contributions to the various impacts, graphs of the process specific impacts are also included for some categories where it is especially relevant. This will better show which processes that are significant in their benefits or burdens. This can help identify any particularly impactful processes in the treatment scenarios, and thus open for more strategic improvements. It is also helpful for the identification of possible limitations of the study.

3 Results and discussion

3.1 Negative carbon emissions

Figures 3.1 to 3.6 show the flows of SS carbon per FU (treatment of 1 ton DM) through the treatment systems and its fate, i.e. how much ends up in the atmosphere and how much goes to a permanent soil storage or is captured by CCS. In the reference case, where conventionally treated SS is applied to land, all of the SS carbon eventually ends up back in the atmosphere, resulting in no negative emission potential.

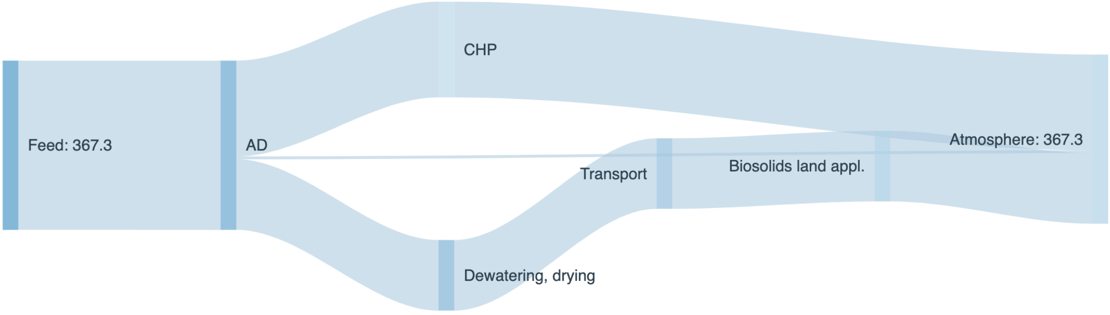


Figure 3.1: Flows (kg) of carbon originating from sewage sludge feed and their fate in the reference case.

The negative emission potential per FU in scenario 1-5 ranges from 37.4 kg C to 226.6 kg C, with scenarios 1 and 2 (AD+PY500) having the lowest and scenario 5 (PY300) having the highest. Note that this negative emission potential only covers the carbon in the SS itself, not any mitigations from fossil fuel replacements. To avoid possible confusion on the parts of the flows that overlap, this overlap was simply generated by the code and does not mean anything for the carbon flows. We see that when AD is included in the treatment pathway, a large amount of the carbon ends up in the biogas. Moreover, it is clear by comparing scenarios 1-2 in figures 3.2-3.3 with scenario 3 in figure 3.4, the increased biochar yield with pyrolysis at 300°C also significantly increases the carbon retained in the biochar, which, combined with a high carbon capture efficiency, increases the negative emission potential from 10% of the carbon to 21%.

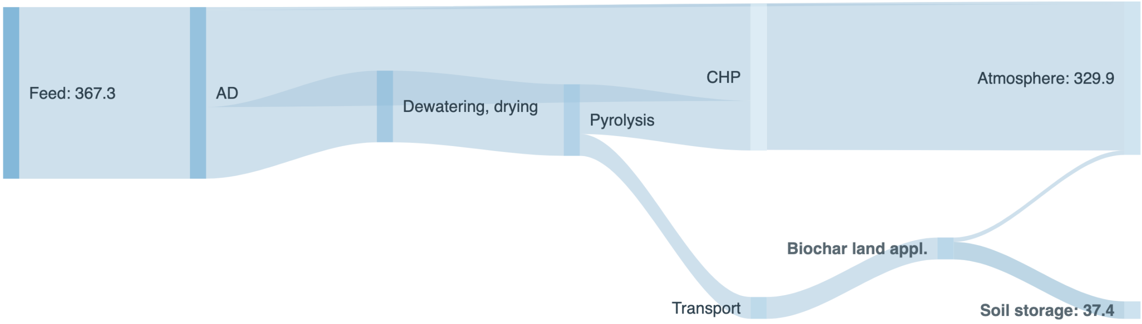


Figure 3.2: Flows of carbon (kg) originating from the SS feed and their fate in scenario 1: AD+PY500, CHP.

When excluding AD from the treatment pathways, the majority of the SS carbon still ends up in the syngas and bio-oil during pyrolysis at 500°C, resulting in a negative emission potential from biochar land application of 29% of the SS carbon, as shown in figure 3.5. The highest negative emission potential is achieved when combining low-temperature pyrolysis with CCS in scenario 5. Due to the high biochar

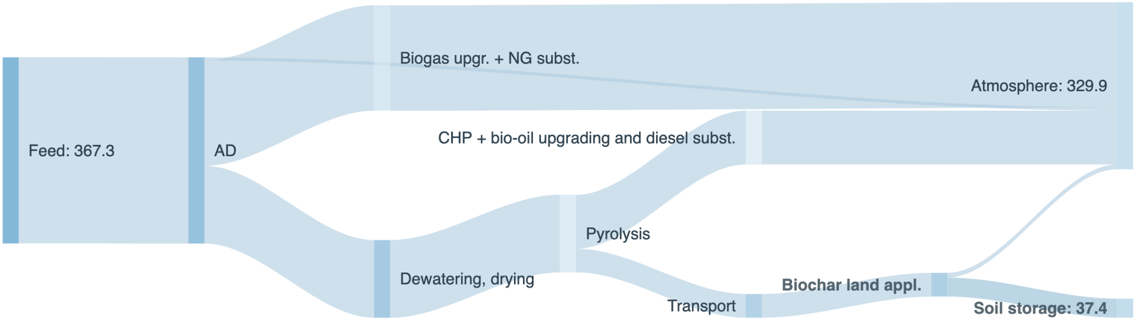


Figure 3.3: Flows of carbon (kg) originating from the SS feed and their fate in scenario 2: AD+PY500, upgrading.

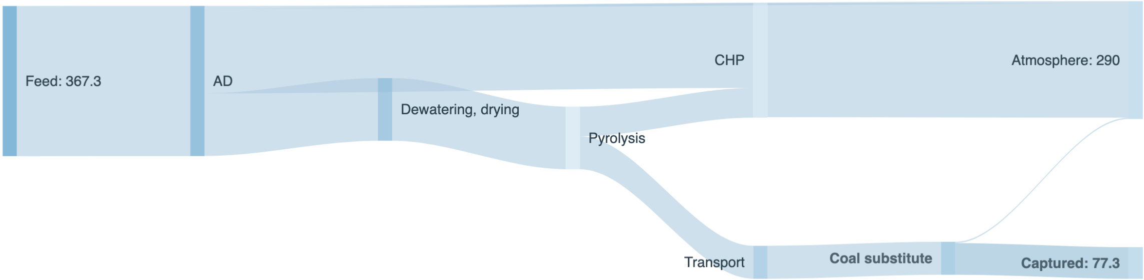


Figure 3.4: Flows of carbon (kg) originating from the SS feed and their fate in scenario 3: AD+PY300.

yield, most of the carbon ends up here, and with a capture efficiency of 98% this gives a negative emission potential of 62% of the SS carbon.

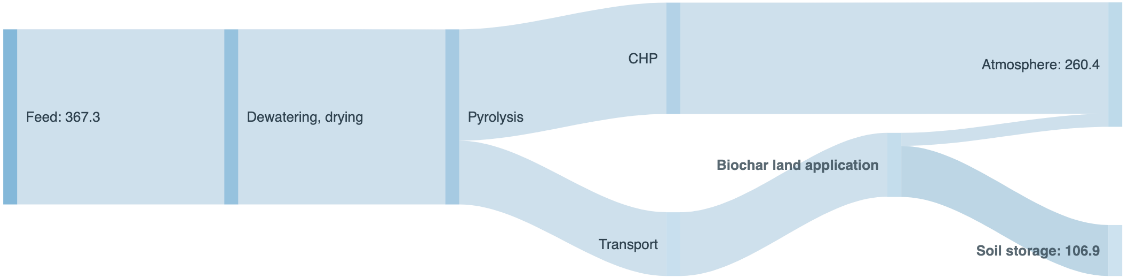


Figure 3.5: Flows of carbon (kg) originating from the SS feed and their fate in scenario 4: PY500

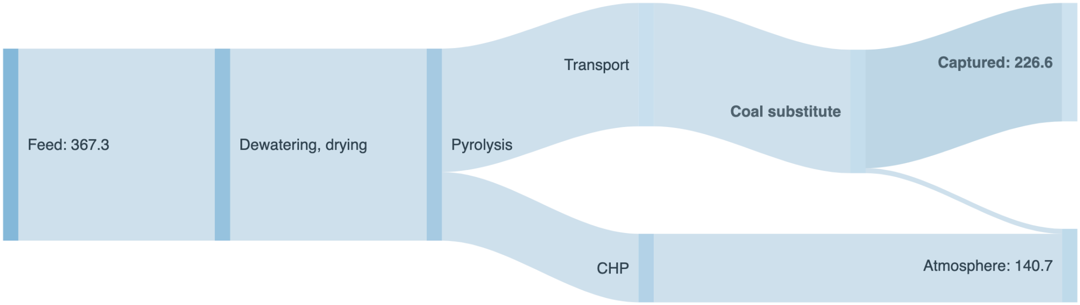


Figure 3.6: Flows of carbon (kg) originating from the SS feed and their fate in scenario 5: PY300

The flows of carbon in the biogas and digested SS were calculated based on the assumptions in table 2.4, and the calculation is shown in table E8 in the Appendix. In addition, the flow of carbon in the bio-oil and syngas is based on the difference between the carbon into the pyrolysis process and the carbon in the biochar. Since the compositions of the pyrolysis products have been gathered from different reference studies, there is not necessarily a carbon balance through this process. The biochar compositions were selected to correspond as much as possible with the process conditions and feedstock compositions, so these are likely more accurate than the compositions of bio-oil and syngas.

Given the average annual SS production of 114 000 tons DM [18], the scenarios' total negative emission potential from Norwegian sludge is summarized in table 3.1.

Table 3.1: Maximum annual negative emissions in the different scenarios, given a Norwegian SS production of 114 000 tons DM per year.

Scenario	**Negative emissions (ton C)**	**Negative emissions (ton CO_2)**
1: AD+PY500, CHP	4264	15 647
2: AD+PY500, upgr.	4264	15 647
3: AD+PY300, CHP	8812	32 340
4: PY500, CHP	12 187	44 726
5: PY300, CHP	16 040	58 867

3.2 HOC removal and HM immobilization

Based on the assumed removal rates and pyrolysis yields, the amounts of the different HOCs that end up in soil, for a given amount of SS treated in the AD+PY500 pathways, are shown in figure 3.7. The table combines the reductions in concentrations after AD, dewatering and pyrolysis with the various mass yields, to find the total mass load of each HOC through the system and being emitted to soil. We can see that for most of the HOCs, the AD can reduce their mass load by 35-80% (although there is uncertainty if this applies in large-scale AD reactors). Therefore, most of the HOCs remain at a similar concentration in the digested sludge compared to before AD. Given the assumed removal rates during pyrolysis, it seems that almost all the HOCs are removed to below detection limit in the biochar. The HOCs that still remain at high concentrations in the biochar are DEHP, PFOS, PFOA (overlapping in figure) and LAS. However, the removal rate of DEHP during pyrolysis was based on low-temperature pyrolysis as no study was found for higher temperatures. Therefore, there is a possibility that it could be removed more than what is assumed here. The removal of LAS was based on a study where the LAS was not in SS, so for this substance as well, the removal during SS pyrolysis at 500°C is uncertain.

As shown in figure 3.8, the remaining amounts of HOCs in the PY500 scenario are quite similar as in AD+PY500. Most of them are reduced to 0-1% of their initial mass during pyrolysis, but for those who are not, quite high amounts remain in the biochar. LAS in particular has a high concentration in the biochar from PY500, as evident from table 3.2. That being said, LAS biodegrades relatively fast under aerobic conditions, giving it a half-life of maximum one week in SS-amended soil [115]. Consequently, there does not seem to be any accumulation in soil nor bioaccumulation in plants, so its high concentration in biochar might not be a major obstacle for land use.

On assignment from the Norwegian Environment Agency, COWI has proposed maximum limits for a selection of HOCs in fertilizer products. Based on the available literature, the concentrations of the selected HOCs in the biochars will be well below the proposed limits. However, since the initial HOC concentrations were averages, it is possible that some SS has concentrations much higher than the ones assumed. In addition, the removal rates assumed for this study might not be constant for any initial concentration; it could be that SS spiked with high amounts of a HOC (as sometimes done in the studies of their removal rates) will result in a higher removal rate than SS with low concentrations of HOCs that are strongly sorbed to the solids.

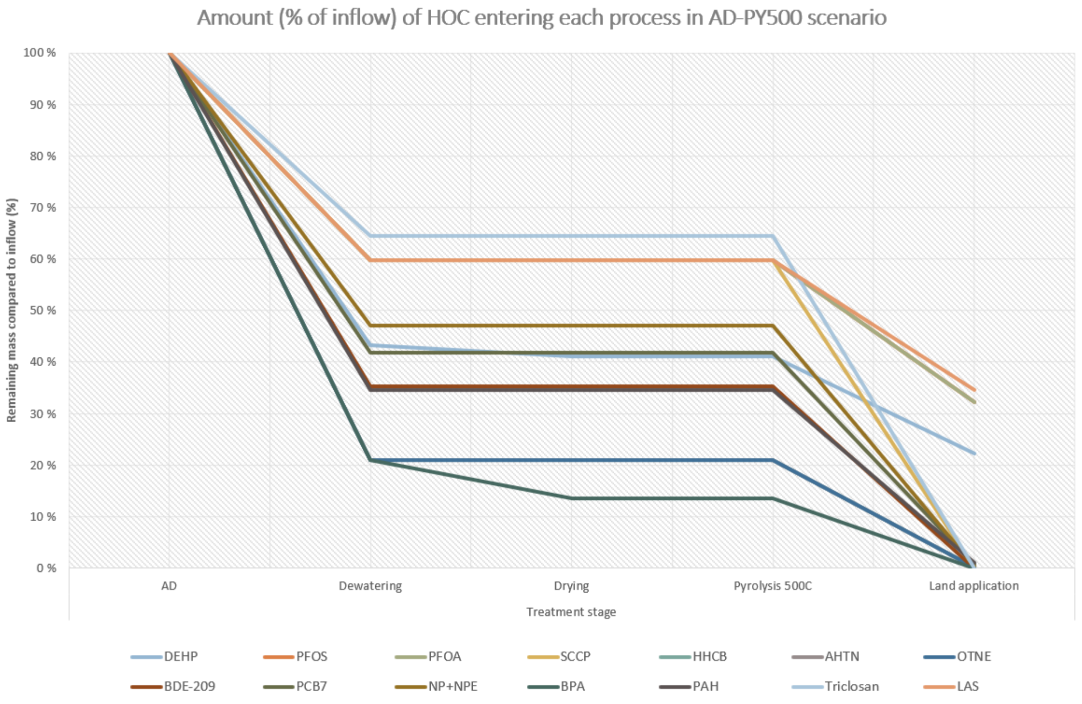


Figure 3.7: Fraction of HOC (% of mass inflow) entering each treatment process and being spread on land in the AD+PY500 scenario.

Table 3.2: Resulting concentrations (mg/kg DM) of the selected HOCs in the biosolids and biochars to be applied on land, plus the proposed maximum limits for Norwegian fertilizer products.

HOC	Ref. case: biosolids	Sc 1 and 2: AD+PY500	Sc 4: PY500	Proposed limit
NP + NPE	3.250	0.033	0.041	10
DEHP	19.01	19.01	26.22	50
BPA	0.3649	0.00	0	-
BDE-209	0.240	0.00	0	0.5
PAH	0.963	0.048	0.083	-
Triclosan	0.640	0.00	0.00	-
PFOS	0.014	0.014	0.014	0.1
PFOA	0.001	0.001	0.001	0.1
SCCP	0.572	0.00	0.00	2
HHCB	3.493	0.035	0.100	10
AHTN	0.875	0.009	0.025	10
OTNE	4.9	0.049	0.140	-
PCB 7	0.012	0.004	0.004	0.02
LAS	14.91	16.02	26.21	-

The HOC results suggest that biochar is a better option for agricultural use than a lot of biosolids used today. The amount of HOCs that is spread on land in the reference case can easily be seen from figure 3.7 after the drying process. While the concentrations of some HOCs are higher in the biochar than the biosolids, we see that their total emissions per FU are still lower. Considering that most of the SS is recycled back to soil in Norway, adding a pyrolysis step to the treatment pathways could significantly reduce emissions to soil of organic contaminants from SS.

Another concern when it comes to SS biochar is that it can contain toxic compounds formed *during* pyrolysis, such as polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs). These compounds were not investigated in this study, but several studies have found that the concentration of PCDD/Fs in SS biochar remains negligible and is expected to have low environmental impacts [116, 117].

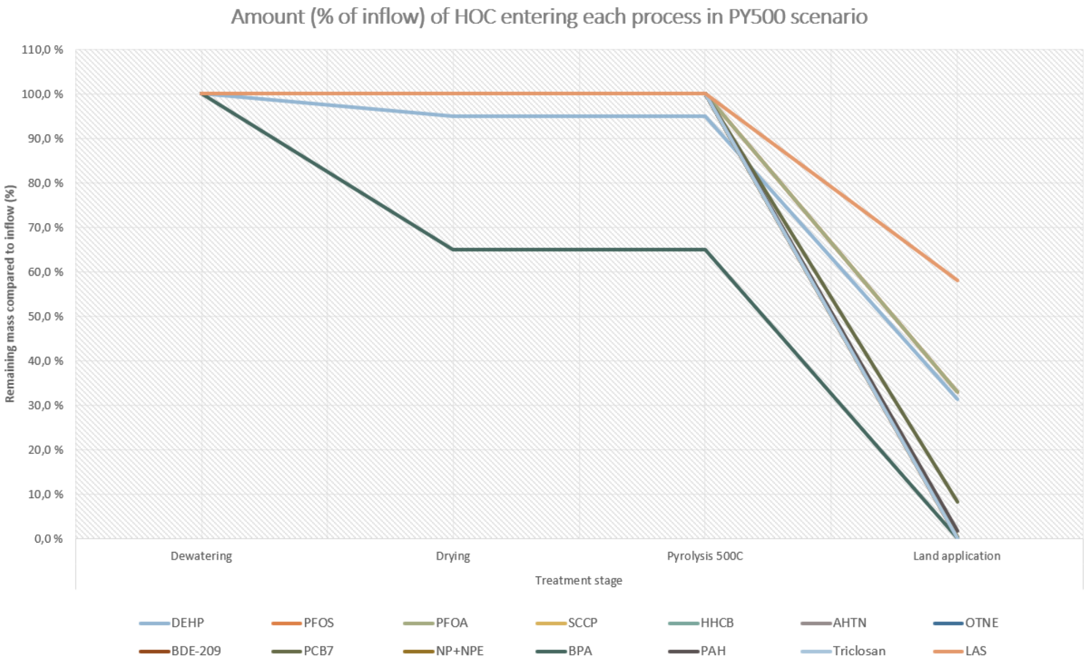


Figure 3.8: Fraction of HOC (% of mass inflow) entering each treatment process and being spread on land in the PY500 scenario.

This study also assessed the fate of HMs in the scenarios to be able to address another concern for SS biochar. In many countries, including Norway, the HM content in organic fertilizers determine its quality class, which in turn determines where and how often it can be applied. The concentrations of HMs in the biosolids and biochars applied on soil are given in table 3.3. With the exception of Hg, the concentrations are approximately twice as high in the biochar as in the biosolids. Most of the HM concentrations in the two biochars are within class I or II, but because of the high concentration in Zn in the biochar it would be classified as quality class III. This class cannot be applied to any area growing food or crops, and thus presents a big obstacle to the application of biochar as a fertilizer substitute and soil amendment. On the other hand, the amount of HM that can be considered bioavailable to plants is still lower per kg biochar than per kg biosolids. By multiplying the concentrations with their bioavailabilities estimated from the findings in Lu et al. [27], we see that there is less available HMs to plants per kg biochar than biosolids. Based on these estimates, the biochar does not seem to have a higher toxicity than the biosolids despite the increased HM concentrations, and could be applied to agricultural soil with less risk of being accumulated in plants. Not only that, biochar has also been proved to be able to reduce the content of available HMs in the soil and plants if applied to already contaminated soil because of its adsorbent effect [118], but the long-term effectiveness is unknown.

Table 3.3: HM concentrations as mg/kg DM in biosolids or biochar in the scenarios with land application. The bioavailable content as mg/kg DM is shown in parentheses.

HM	Ref. case	Sc 1 and 2: AD+PY500	Sc 4: PY500
Pb	22.0 (0.44)	38.7 (0.37)	37.8 (0.37)
Zn	615.1 (87.87)	1119.7 (75.8)	1093.5 (74.0)
Ni	23.0 (1.37)	39.9 (0.80)	38.9 (0.82)
Cd	1.0 (0.05)	1.8 (0.02)	1.8 (0.02)
Cu	284.0 (14.8)	457.1 (0.45)	446.4 (0.44)
Cr	30.8 (1.3)	46.0 (0.58)	44.9 (0.57)
Hg	0.67 (0.003)	0.00	0.00

3.3 Life cycle assessment

Figure 3.9 and 3.10 show the net impacts of the reference case and the 5 scenarios in the selected midpoint categories from ReCiPe and USEtox. For comparative purposes, the results were scaled in each category based on the scenario with the highest impact in absolute value. The highest or lowest impact was given the value 1 or -1 respectively, and then the other impacts were scaled to values between 1 and -1 relative to this. A low value indicates less negative environmental impacts. Negative values occur when the avoided products or emissions outweigh the impacts associated with the operation of the systems themselves. This can be misleading, as it suggests that the systems can have a positive effect on the environment when in reality they need heat, electricity and material for their operation, which would have to come from other sources if it was not for the internal CHP system. It is thus the system expansion done to capture the benefits of the co-products that brings down the net impacts and sometimes results in negative values.

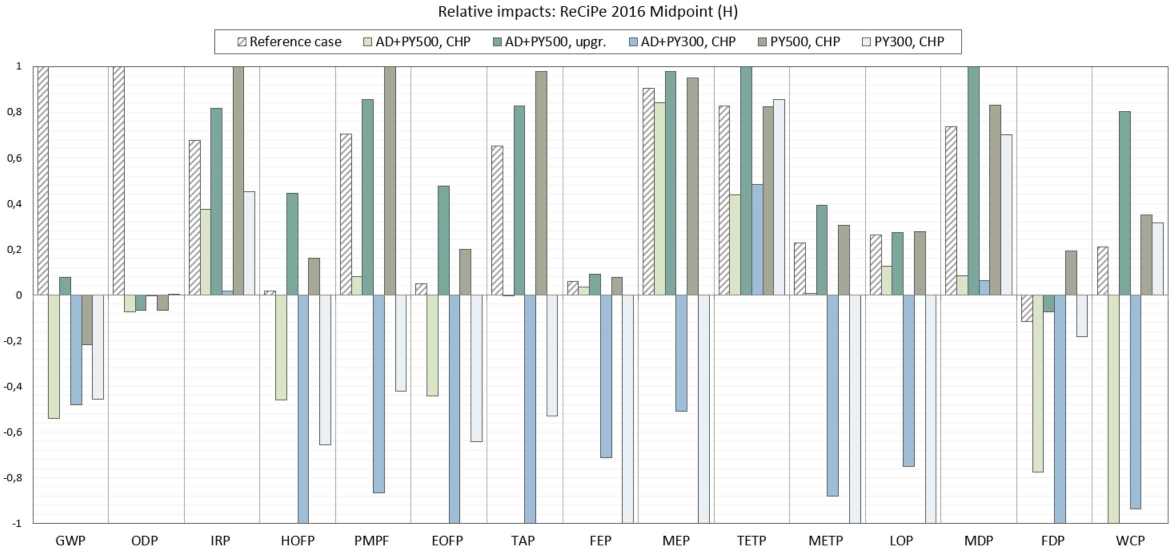


Figure 3.9: Relative LCA impacts for the categories used from ReCiPe Midpoint 2016 (H).

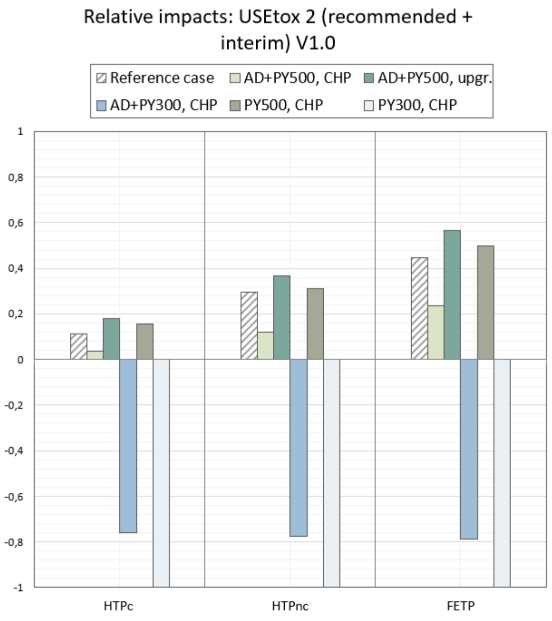


Figure 3.10: Relative LCA impacts for the categories used from USEtox 2 (recommended + interim) V1.0.

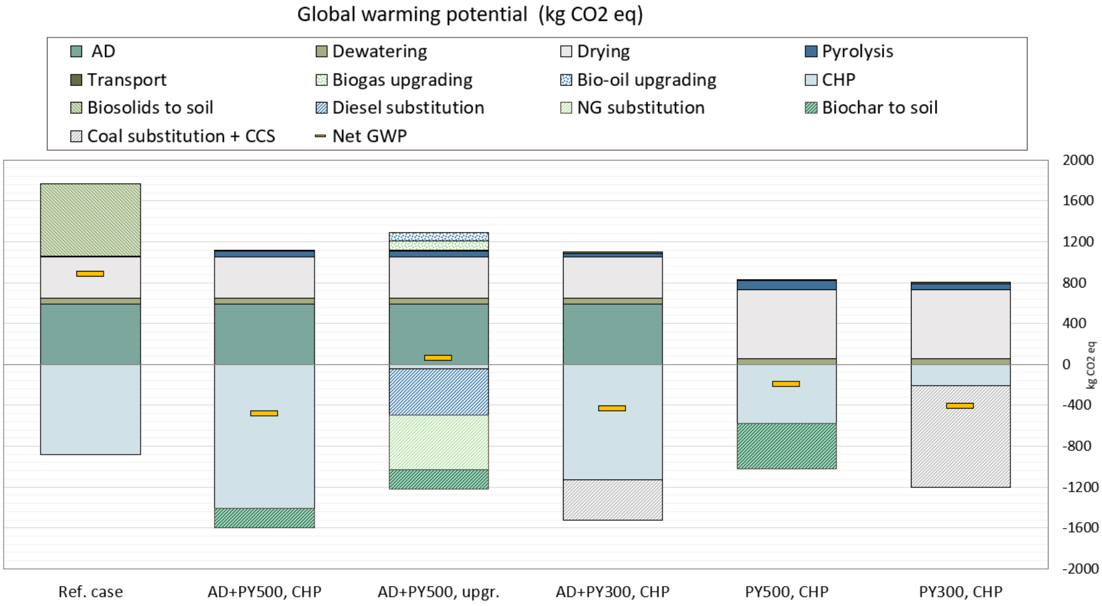


Figure 3.11: Process contributions to GWP and the net GWP of each scenario.

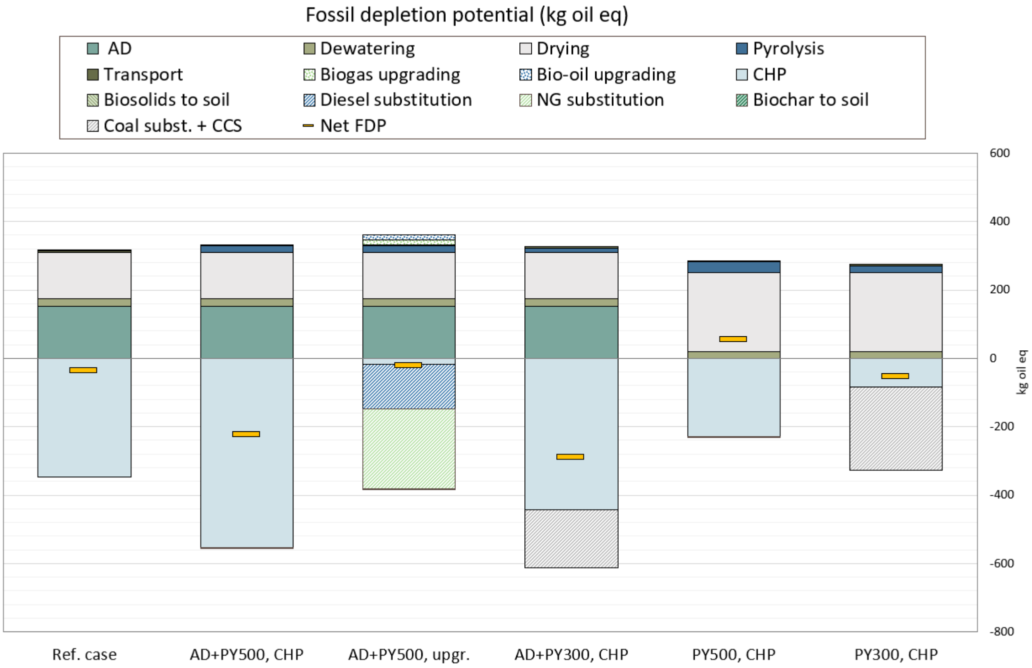


Figure 3.12: Process contributions to FDP and the net FDP of each scenario.

The reference case for Norwegian SS treatment and disposal is usually in mid- or upper-range in most categories. The biogas from AD and lack of energy input to pyrolysis gives it an advantage in some categories, such as ozone formation in HOFP and EOFP as well as the fossil depletion potential, FDP. We see that the CHP of the biogas in the reference case outweighs the impacts from the AD. However, it rarely achieves net negative impacts since it has limited fossil fuel substitution potential. Moreover, the N_2O emissions from the land application of biosolids significantly increases the net GWP - and that is just from 2.37 kg N_2O per FU. These emissions are therefore included in a sensitivity analysis in the next section since they have a significant uncertainty. From figure 3.11, we can see that nearly all the avoided burden from the CHP of the biogas is lost because of the N_2O emissions from land application. This

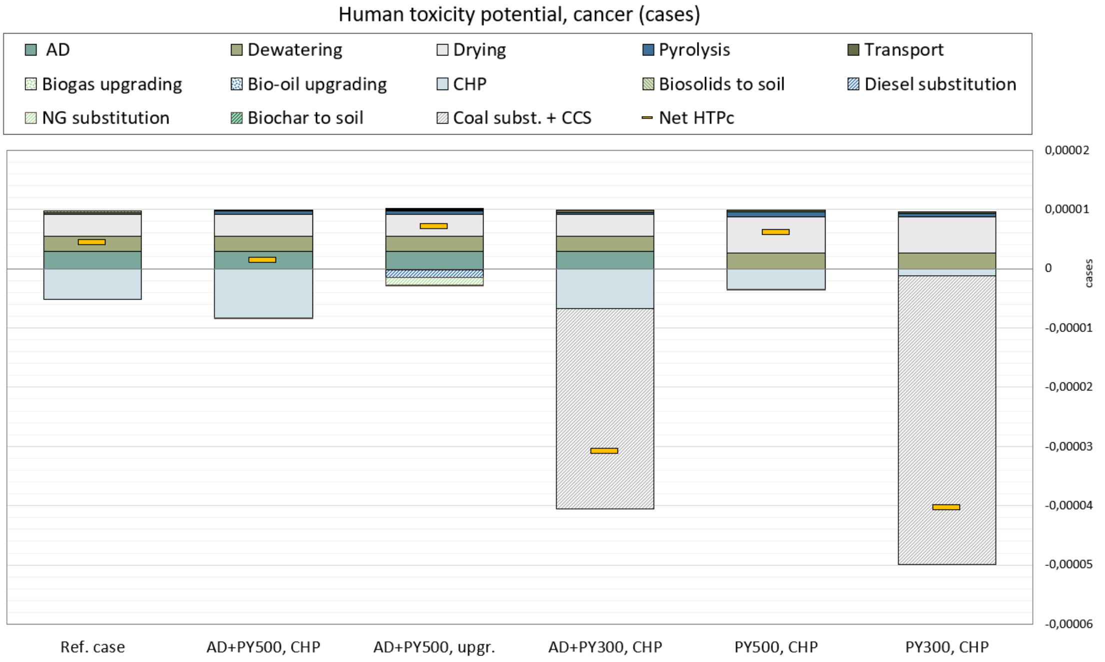


Figure 3.13: Process contributions to HTPc and the net HTPc of each scenario.

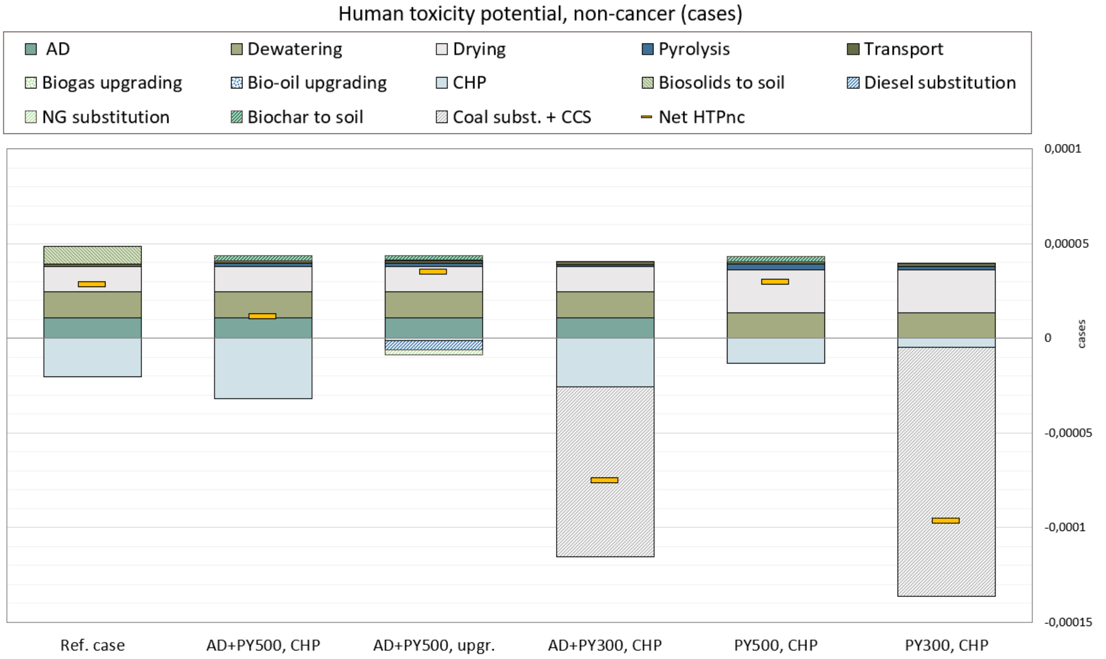


Figure 3.14: Process contributions to HTPnc and the net HTPnc of each scenario.

does not affect the FDP however, so for this category, the reference case performs better than PY500, which has the highest FDP due to the relatively low fossil fuel substitution potential and increased energy demand for drying.

The main disadvantage for pyrolysis of SS is the large heat requirements for the SS drying, and to some extent the heat for the pyrolysis process itself. The CHP of only syngas and bio-oil in PY500 and PY300 is not enough to outweigh their GWP contributions. If fossil fuels are used for the generation of this heat, and not the pyrolysis products or other renewable sources, this would reduce the benefits of pyrolysis substantially. It is therefore paramount to take advantage of the pyrolysis products to counteract the

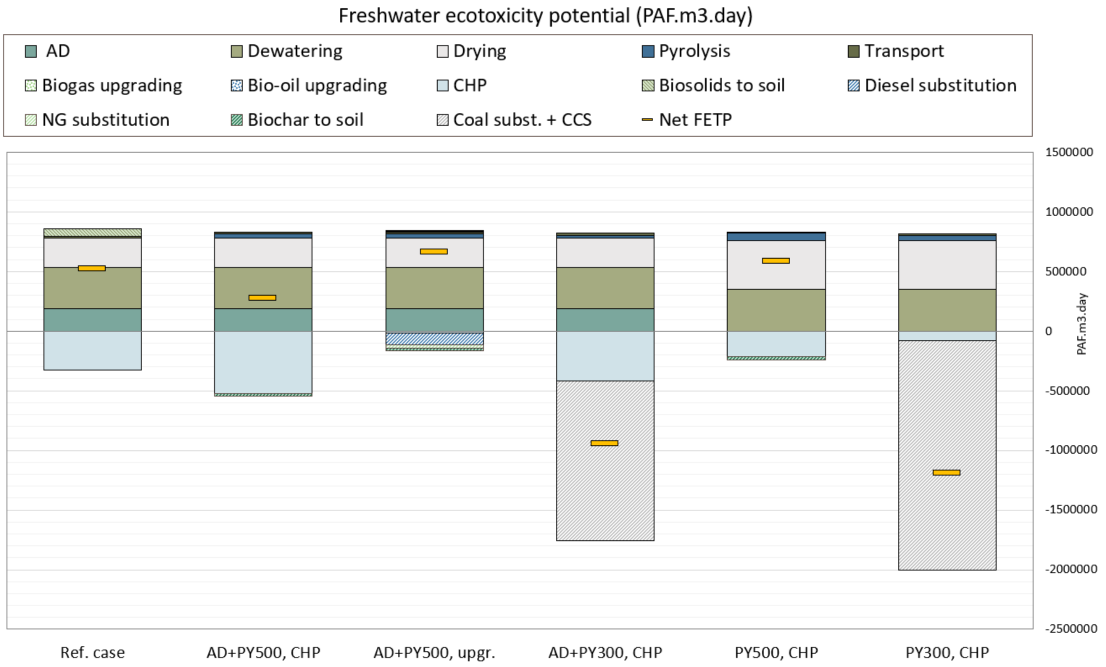


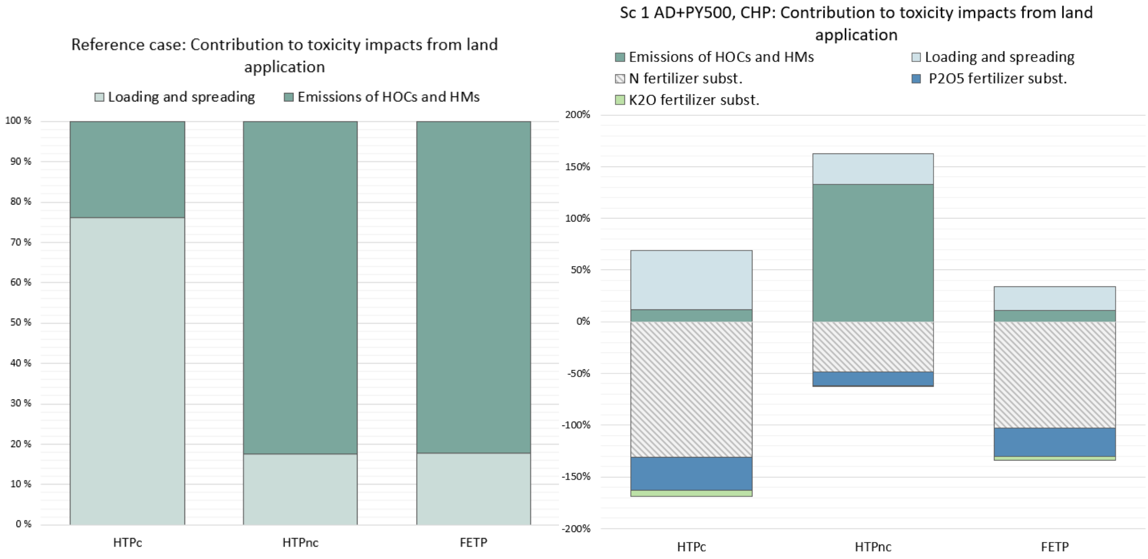
Figure 3.15: Process contributions to FETP and the net FETP of each scenario.

additional impacts from drying and pyrolysis. Using the excess heat from pyrolysis to dry the SS is also an option to reduce the need of fossil fuels. This is also why the GWP is reduced if the pyrolysis is combined with AD, since the biogas can supply a lot of the energy demand with no GHG emissions (except potential methane losses) due to the biogenic origin of the SS carbon. Upgrading of the biogas to biomethane is also an option, but since the systems with pyrolysis have a higher energy demand than the bio-oil and syngas can substitute, it is likely better to use the biogas to supply this energy unless there are other renewable alternatives. The same goes for upgrading of bio-oil as it requires both natural gas and electricity. The GWP is reduced if it is instead used in a CHP to substitute natural gas.

We can see that the scenarios with coal substitution, AD+PY300 and PY300, have low impacts in most categories due to the many negative effects associated with coal mining and use. In some categories, HOFP, EOFP, TAP and FDP, the biogas production in AD+PY300 is more important than the added negative emissions and coal substitution in PY300. However, for nearly all the categories related to eutrophication and toxicity, PY300 comes out as the best choice. The effect of coal substitution is even more evident in the figures 3.11 to 3.15, where the contribution from coal substitution and CCS is shown more clearly. While most processes have a net positive impact (i.e. negative for the environment), "Coal substitution + CCS" brings the net impacts down for all selected categories. It is particularly dominating in the toxicity impact categories, whereas for the GWP and FDP, the effect of the CHP is more significant in most scenarios. The same effect is clear in figures 3.13 to 3.15 - the CHP in scenario 1 has a consistently higher benefit than the upgrading to fuels.

In contrast to the results for the negative emission potential, scenarios using AD and pyrolysis combined with CHP comes out with the best climate impacts in terms of their GWP. This is in line with results from other studies comparing similar scenarios [21, 22, 32]. Scenario 2, where the biogas and bio-oil is upgraded to replace NG and diesel, does not get the as high avoided burdens as scenario 1 using CHP. This could have been different if other methodological choices were made for the upgrading scenario, such as substituting the combustion of diesel in agricultural machinery instead of just the CO_2 emissions from it. This was attempted initially, but gave an unrealistically high avoided impact to the toxicity categories because of all avoided emissions from the life cycle and operation of the machinery. Most likely, the combustion of bio-oil will release toxic compounds similar to that of diesel, particularly since some of the toxins in the SS volatilize and might end up in the oil. The positive effect of bio-oil and biogas upgrading and fuel substitution is also clear in figure 3.12, but even here it is not as beneficial as the substitution of heat and electricity in the CHP. If it was instead assumed that the electricity and heat consumed in the systems were from less carbon-intensive sources, such as hydro, the positive effect from CHP would likely have been a lot smaller, since. as mentioned, all processes are supplied by energy from a CHP running on NG. For the USEtox impacts, the benefit of CHP becomes less dominating and the coal substitution gives scenario 3 and 5 clearly lowest toxicity impacts.

Something worth noting is that the toxicity impacts from soil application of biosolids and biochar are so small that their contributions are barely visible in figure 3.13 and 3.15. For the total impacts to HTPnc and FETP in the reference case, we can see that the soil application has a small but noticeable impact. In the other scenarios with soil application, the impacts from this process are much lower and even negative for FETP. However, there are other reasons to this than just the destruction of HOCs and immobilization of HMs; firstly, the spreading of biochar requires less diesel etc. per FU than the spreading of biosolids due to less biochar being produced than biosolids (see table F1 for the amounts). In addition, the substitution of synthetic fertilizers, even though very little, also contributes to reduce the impacts from the soil application process, resulting in net negative impacts in some scenarios. To illustrate this, the contributions to the toxicity impacts from soil application in the reference case and in scenario 1 are shown in figure 3.16. In the reference case, the emissions of HOCs and HMs in the categories HTPnc and FETP account for more than 80% of the impacts, whereas the spreading process dominates the HTPc impacts. This is not surprising considering that almost no HOCs had characterization factors for the HTPc category, as shown in table B1 in the Appendix. With the fertilizer substitution in scenario 1, the substitution of N fertilizer is what contributes the most to bring down the impacts in HTPc and FETP. The relative contribution from HOCs and HMs are much smaller in the FETP category in scenario 1, indicating that the reduction in HOCs and HMs has an effect in reducing the FETP impacts.



(a) Reference case.

(b) Scenario 1: AD+PY500, CHP.

Figure 3.16: Process contributions (% of total) land application of biosolids in the reference case and scenario 1: AD+PY500, CHP.

To identify exactly which HOC and HM compounds contribute the most to toxicity impacts in soil application of biosolids and biochar, the contributions were made relative as shown in figure 3.17 and 3.18. It is very interesting to see that the chosen priority HOCs contribute almost nothing to the impacts in HTPc, HTPnc and FETP compared to the HMs. Though, this is also because their emissions per FU are lower than the HM emissions by several factors, as shown in table F1. In the HTPc category, Ni contributes to 94.7% of the impacts while the sum of PAH16 accounts for only 4.3%. Zn is responsible for 98.2%

of the impacts in HTPnc and 7.6% of the FETP impacts, with Cu comprising the remaining 92%. Circling back to the change in the HMs bioavailability after slow pyrolysis at 500°C, as shown in table 2.11, it is reduced from 5.97% to 2.10% for Ni, from 14.29% to 6.77% for Zn and from 5.22% to 0.10% in Cu. If these estimates, based on experimental findings in Lu et al. [27], are also valid in actual agricultural soil conditions, this is promising for the agricultural use of SS biochar as an alternative to biosolids. The large decrease in bioavailability of Cu is evident in the FETP category in figure 3.18, where now Zn dominates the impacts instead.

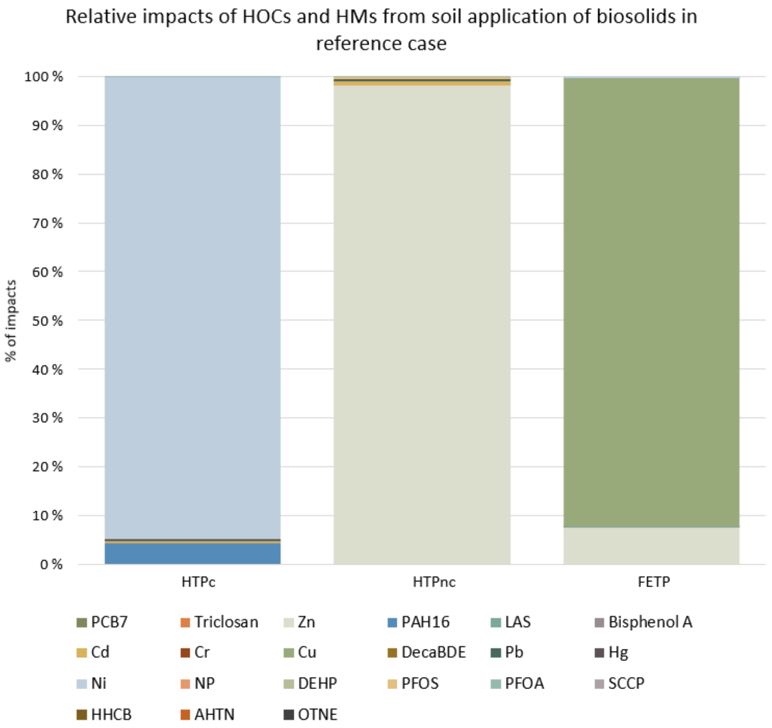


Figure 3.17: Relative impacts of the included HMs and HOCs in SS applied to soils in the reference case.

Furthermore, the results in figure 3.17 and 3.18 are for only the bioavailable emissions; their actual emissions are much higher and would dominate even more in the toxicity impacts. Something worth mentioning, however, is that SS only accounts for a small fraction of the total amount of HMs that is added to soils in Norway, as the majority comes from atmospheric deposition and synthetic fertilizer [119]. However, it is also relevant to mention that Zn and Cu are essential micro-nutrients for both plants, humans and animals, and it is only in too high amounts that they become toxic. Similarly, Ni and Cr can be important for some biological processes [119].

Given the current levels of HOCs and HMs in SS, it is clear that HMs, and in particular Ni, Zn and Cu, are a significant contributor to human toxicity and freshwater ecotoxicity when the SS is applied to soils. Of course, if this study had included all the hundreds of organic contaminants and medicinal residues present in SS, figure 3.17 might have looked very different. For any further studies, it would be interesting to see exactly how much the sum of all organic contaminants etc. contribute to the toxic impacts of soil application. Here, only a selection was included in order to carefully assess their fate during pyrolysis, thus the net impact of organic contaminants in SS applied to soils is still not determined.

Lastly, it is also relevant to mention that the feasibility of the scenarios, both in terms of economic and technical aspects, are not included in this analysis. Even though the results show clear advantages to coal substitution, this might be very challenging to implement in actual cement kilns or other industries. In addition, the use of commercial-scale CCS in industries is not yet very common due to the high investment requirements. Until carbon tax or other incentives motivates the use of biochar to substitute

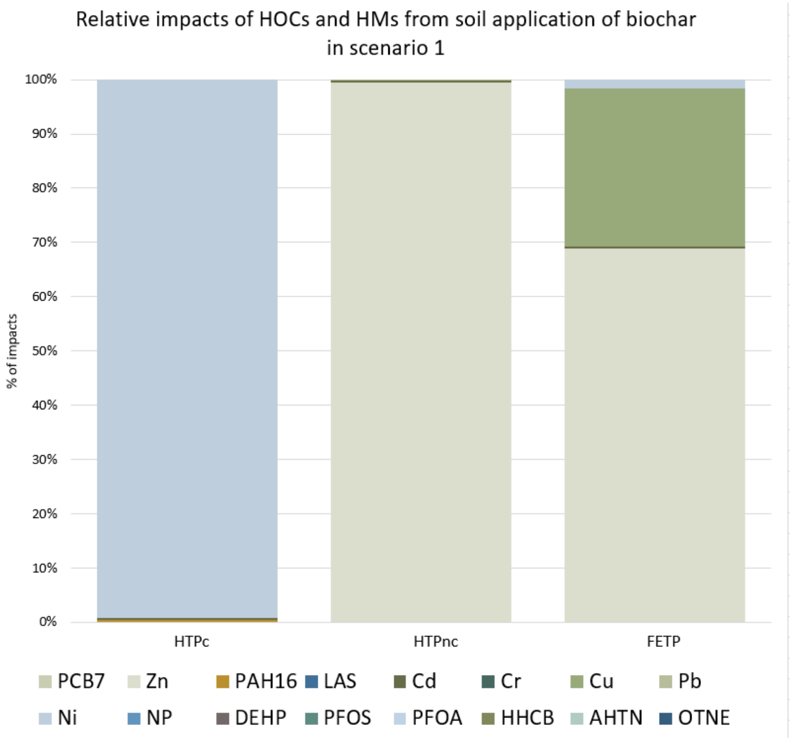


Figure 3.18: Relative impacts of the HMs and HOCs applied to soils from biochar in scenario 1.

solid fuels, the scenarios with coal substitution and CCS are probably the least realistic ones. Another downside with the use of biochar as a solid fuel is that one loses the nutrient recovery potential. The world may experience a shortage of chemical P fertilizers in the future, increasing the importance of improving P recovery in sectors such as here [120].

4 Sensitivity analysis

4.1 Variations of process parameters

Specific biogas production
The production of biogas during AD will usually be in the range 0,9 - 1,1 m^3/kg VS destroyed [31]. Since the biogas contributes a lot to the reduction of GWP and FDP when used in a CHP or as a natural gas substitute, a sensitivity analysis was done on the effect on the LCA impacts of changing the biogas production of +-10% from the original assumption.

Biochar yield
Because the biochar yield is so dependent on process conditions and feedstock properties, it has a larger uncertainty than the biogas production. Based on studies with similar process conditions (temperature and residence time) and feedstocks (SS with similar pre-treatment) as used in this study, it seems that the biochar yield can still vary around +-15% from the yield assumed in this study [9, 56, 60, 65]. The sensitivity analysis will attempt to capture the effects of this uncertainty on the LCA impacts. The change to the biochar yield caused a change to the yields of the other products in accordance to their initial yield to get a sum of 100%. See tables E4 to E7 in Appendix E for details.

Biochar carbon content
The carbon content in the biochar is a key parameter for the negative emissions in the scenarios investigated. This value will also be uncertain since it depends on the same factors as for the biochar yield. By reviewing studies with similar treatment pathways and feedstocks, it was found that the carbon content usually lies in the range 14-18% for digested sludge [60, 65], so the same uncertainty is assumed for undigested sludge, which is approximately +-20% of that used in this study.

Transport of biochar and biosolids
Since the original transport distance of biosolids and biochar was chosen somewhat haphazardly, a sensitivity analysis of changing the distance +-30% is included to see if this has a significant impact on the LCA results.

Impact on soil N_2O emissions from biosolids and biochar application to land
For the N_2O emissions caused by biosolids application to soil, the uncertainties as mentioned in IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories [6] for the direct and indirect emissions were used: 25% uncertainty in direct N_2O emissions and 50% uncertainty in the emission factors and fractions used for indirect emissions. This resulted in the N_2O emissions changing by +-47% in the reference case.

The sensitivity analysis on the reduction in N_2O emissions due to land application of biochar was done using the uncertainty ranges in table 2.12, while keeping the assumed fertilizer substitution constant. Using the upper and lower values changed the initial N_2O emission reductions by approximately +-30%.

4.2 Results and discussion of sensitivity analysis

The results of the sensitivity analysis for impacts in the GWP, FDP, HTPc, HTPnc and FETP categories are shown in figure 4.1 to 4.4. Note that the change in N_2O is different in the reference case, where it is +-47% emissions, while in the other scenarios it changes with +-30%. Moreover, since for scenarios 1-5 it is a sensitivity in the *effect* of biochar on N_2O emissions, a negative change in the figure means less reduction in the emissions because of biochar application, i.e. an increase in N_2O and vice versa.

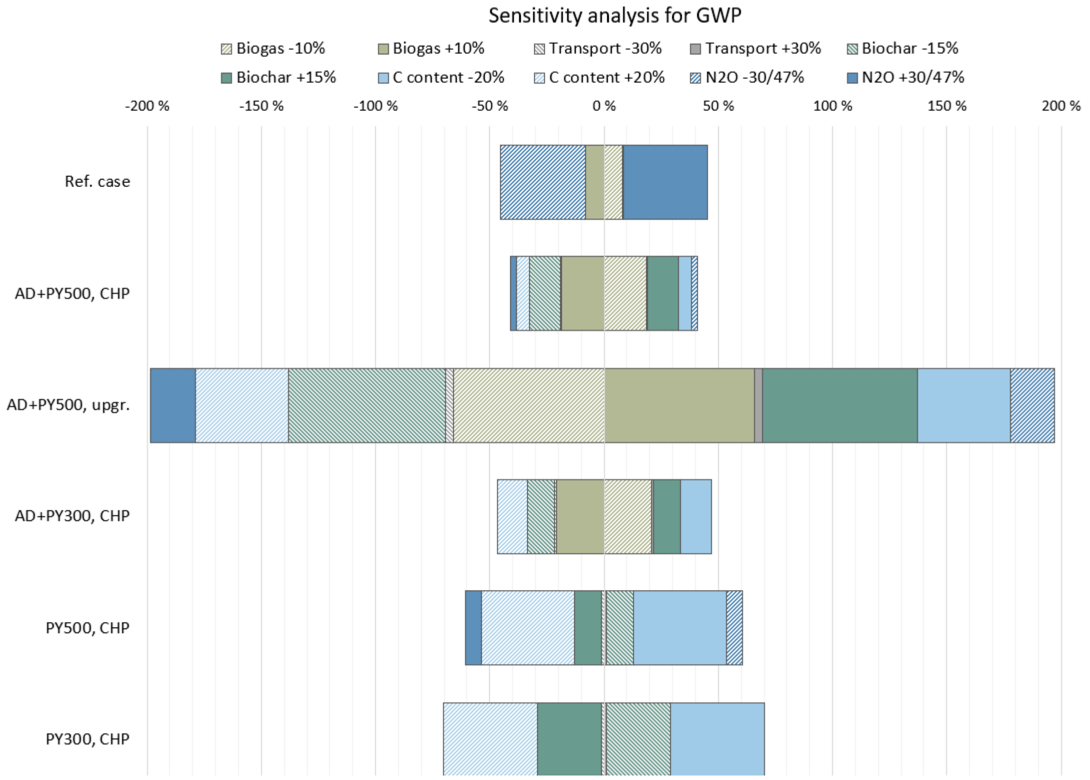


Figure 4.1: Sensitivity of GWP impacts (% change from the scenario's original GWP) to the changes in the selected variables.

As shown in figure 4.1, the variation in N_2O emissions is clearly what has the most significant effect in the GWP of the reference case. A reduction of 47% in the estimated N_2O emissions from SS land application would reduce the net GWP for the system by 37%. The uncertainty in the biogas production only results in a change of +-8% in the GWP, where an increased biogas production naturally reduces the GWP since it substitutes heat and electricity from NG. The uncertainty in transport distance of biosolids has nearly no effect at all on the GWP, with less than 1% change in GWP from a change of 30% in transport distance.

In scenario 1 (AD+PY500, CHP), 2 (AD+PY500, upgr.) and 3 (AD+PY300, CHP), the main uncertainty for the GWP is in fact the biogas production. The variables with the second and third most significant uncertainty is the biochar yield and its carbon content. If the biochar yield increases by 15%, the GWP would increase as a result of the decreased yield of bio-oil and syngas going to the internal CHP, despite the added benefits from soil application of biochar. A change in the biochar's carbon content of 20% leads to a relatively small change in the GWP, but would of course change the negative emission potential. The N_2O emission uncertainty from land application of the biochar has the second smallest contribution in scenario 1 and 2. The transport uncertainty is of very little significance in the GWP for all scenarios.

The two remaining scenarios, PY500 and PY300, all have a relatively high impact from the uncertainty in the biochar carbon content to their GWP, followed by the biochar yield. In these two scenarios, unlike the three previous, an increased biochar yield would decrease the GWP. This is not very surprising since the biochars substitutes coal and 98% of its carbon is assumed to be captured, whereas the syngas and bio-oil simply substitutes NG used in a CHP system. The transport uncertainty is insignificant here as well.

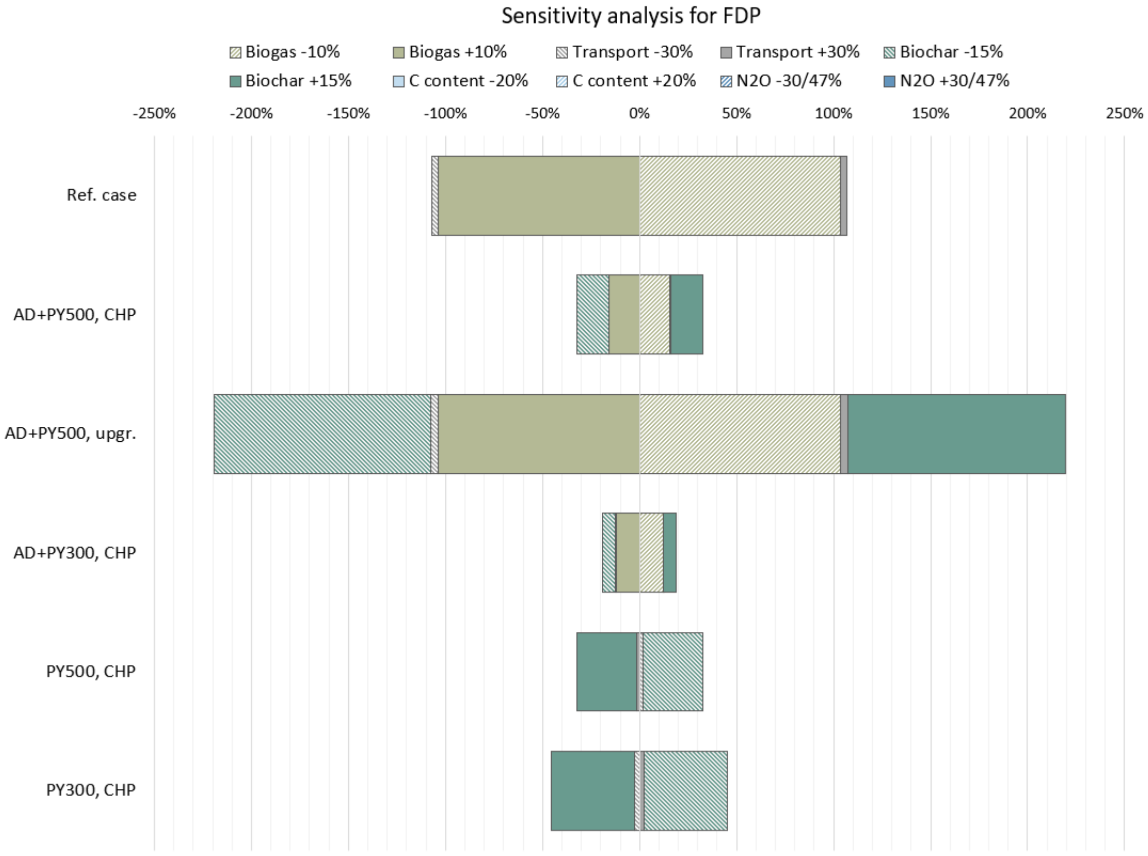


Figure 4.2: Sensitivity of FDP impacts (% change from the scenario's original FDP) to the changes in the selected variables.

As for the FDP, the uncertainty in the biogas and biochar yield have very similar impacts for the AD+PY500 scenarios, while for the AD+PY300 the biochar matters less. Surprisingly, even in scenario 3 with coal substitution, an increased biochar yield would increase the FDP of the system

An increased biochar yield increases the HTPc, HTPnc and FETP impacts for scenario 1, 2 and 4, as shown in figure 4.3, 4.4 and 4.5. In other words, only in the scenarios with coal substitution with biochar are the toxicity impacts reduced with an increased biochar production. This is not strange since an increased yield of bio-oil and syngas benefits increases the substituted heat and electricity from CHP, whereas more biochar in these scenarios only increases the emissions of HOCs (not HMs because of a constant retention rate assumed) and negative emissions of CO_2 and N_2O.

To summarize some key points from the sensitivity analysis, the main uncertainty to the GWP in the reference case is its estimated N_2O emissions from soil application of biosolids. It is therefore unfortunate that no country-specific factors were available for the calculation of these emissions, because it might very well be that they are significantly different from the IPCC's default factors. For the GWP in the other scenarios, the uncertainty in N_2O emission reduction was not as important as the uncertainties in biogas production and biochar yield. Their uncertainties each gave more than three times as high impacts to the GWP than the uncertainty for the N_2O did. The biochar C content was less important than the biochar yield in the scenarios with biochar soil application, but in the scenarios with coal substitution its uncertainty was the most significant one for the GWP. For any further studies, it would therefore be interesting to get more data on SS compositions in Norway, since the biochar carbon content depends on that of the feedstock. That being said, a 20% change to the biochar C content does not have a huge impact on the net GWP of the systems with soil application, so a SS composition specific for Norway would, in all likelihood, lead to the similar results. The toxicity impacts HTPc, HTPnc and

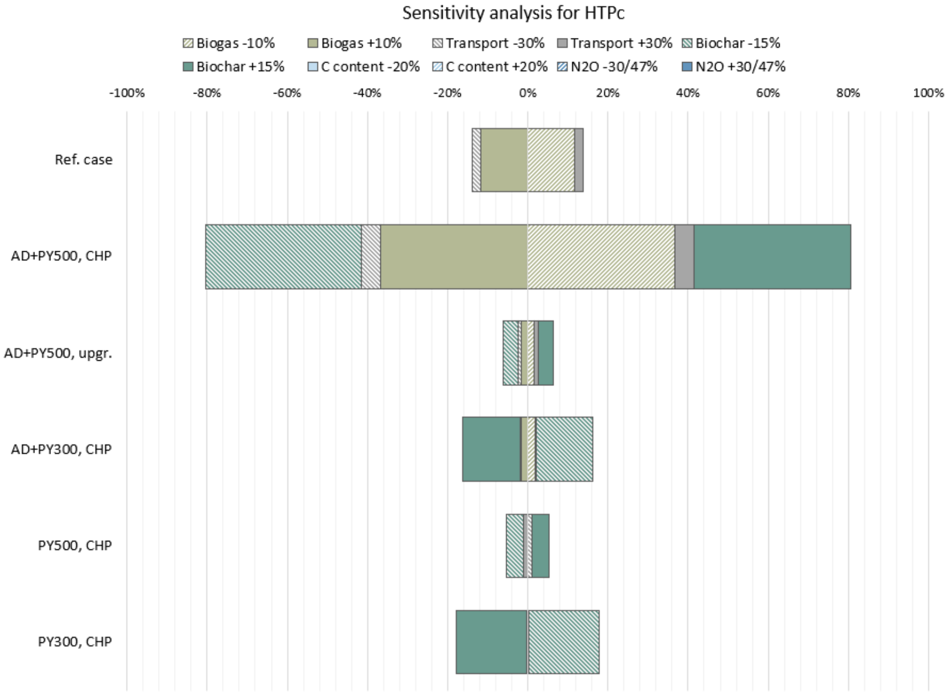


Figure 4.3: Sensitivity of HTPc impacts (% change from the scenario's original HTPc) to the changes in the selected variables.

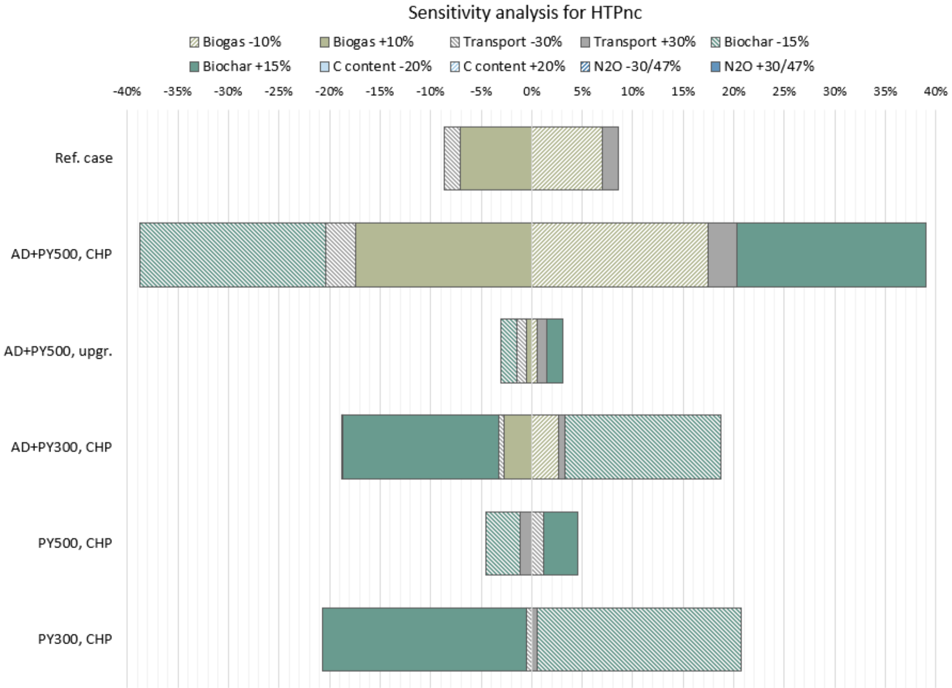


Figure 4.4: Sensitivity of HTPnc impacts (% change from the scenario's original HTPnc) to the changes in the selected variables.

FETP were mostly impacted by the parameters related to CHP substitution, i.e. the biogas and biochar yield. Unless the biochar can substitute coal, the toxicity impacts are reduced by also minimizing the biochar production so that as much heat and electricity as possible can be substituted by the bio-oil and syngas. In addition, it was found that uncertainty in the transport distance of biosolids and biochar is insignificant to all the categories analyzed here.

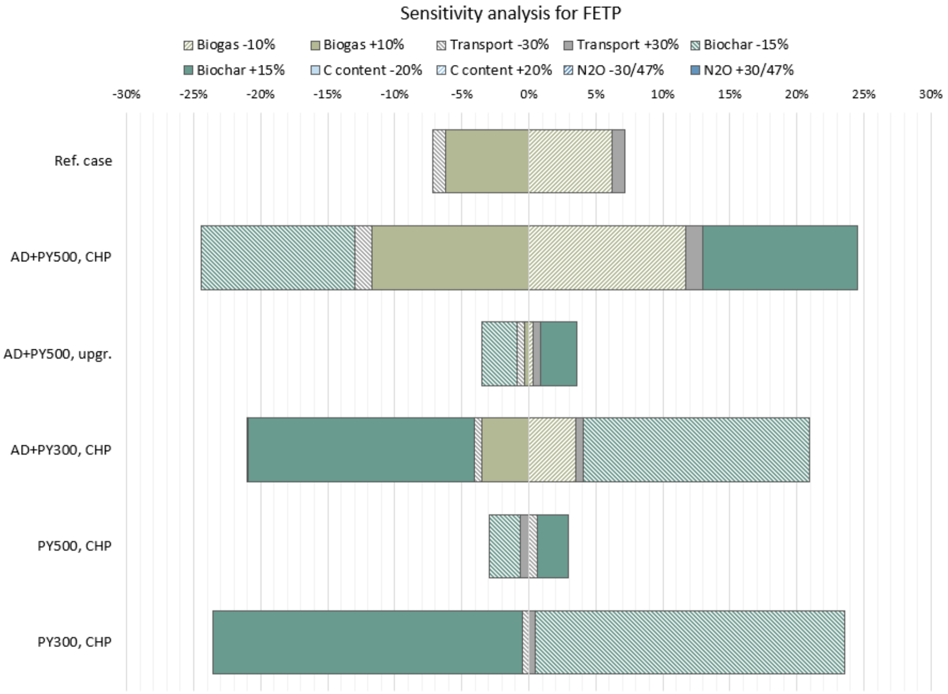


Figure 4.5: Sensitivity of FETP impacts (% change from the scenario's original FETP) to the changes in the selected variables.

5 Conclusion

In this thesis, the environmental impacts of alternative sewage sludge (SS) treatment scenarios for implementation in Norway have been studied by performing a comparative LCA using data from the literature. The alternative SS scenarios include the use of pyrolysis for the abatement of organic contaminants (HOCs) and the production of biochar, whereas the reference case was assumed to be anaerobic digestion (AD) and drying of the SS before agricultural application of the remaining biosolids. Five scenarios with different variations of slow pyrolysis, either integrated with AD or not, were then compared to the reference case. In addition to their total life cycle impacts, the fate of a selection of priority HOCs and heavy metals (HMs) was carefully studied for each scenario to see if pyrolysis can be a method to reduce the toxicity of SS. A detailed quantification of the negative carbon emissions of each scenario was also performed.

The scenarios with pyrolysis had several obvious advantages over the reference case. First of all, the pyrolysis converts part of the SS to bio-oil and syngas which can be used as bioenergy products to substitute the use of fossil fuels. In addition, soil application of biochar does not emit GHGs like the application of biosolids does, instead it can potentially reduce them. The biochar also offers a negative emission potential when applied to soil, or alternatively it may be a possible use as a solid fuel to substitute coal. It was found that the application of biosolids to soil contributes significantly to the GWP of the reference case, and also resulted in higher emissions of HOCs and more bioavailable HMs than the alternative scenarios with pyrolysis. The scenario with the lowest climate impacts in terms of GWP was a combination of AD and slow pyrolysis at 500°C, followed by soil application of biochar for negative emissions. The combination of AD and pyrolysis at 300°C followed by using the biochar to substitute coal resulted in much higher negative emissions, but less fossil fuel substitution potential and thus not as low GWP. Similarly, using only pyrolysis at 300°C gave the highest negative emission potential of 62% of the carbon contained in the initial SS. However, since this scenario attempted to maximize the negative emissions from biochar it had almost no other co-products to substitute fossil fuels, which gave a GWP slightly higher than the two previously mentioned. It is therefore clear that there is a trade-off between the negative emissions and the production of bioenergy products. Whether negative emissions or bioenergy production should be prioritized depends on the accessibility of renewable fuels for the production of energy in that specific case. If heat and electricity to the systems can easily be supplied from renewable sources, perhaps it should be prioritized to avoid using AD and instead maximize the negative emission potential of the biochar.

In terms of the HOCs in SS and the effect of pyrolysis, it was found that slow pyrolysis at medium temperatures (500°C) is a promising technique to destroy many of the priority HOCs included in this study. While higher temperatures might contribute to an increased destruction of some HOCs, it may also result in an increase of other toxic compounds generated at higher temperatures. In addition, a higher temperature significantly reduces the biochar yield and negative emission potential, while also potentially reducing the positive effect on crop yields. As for the heavy metals (HMs), most of them will accumulate in the biochar and have twice as high concentrations as in the biosolids. This is a big obstacle for the use of biochar as a soil amendment and fertilizer substitute, since it would be classified as a fertilizer that cannot be used on any food crops in Norway. Interestingly however, it was found that the pyrolysis reduced the bioavailability of the HMs to such an extent that the bioavailable amounts were smaller in the biochar compared to the biosolids. These results indicate that SS-derived biochar is a better option for agricultural use than the biosolids used today, both in terms of the climate benefits and the toxicity impacts.

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Appendix

A Product compositions

Table A1: Composition of the bio-oils.

	AD+PY300	**AD+PY500**	**PY300**	**PY500**
Ultimate composition (wt. %)				
Ash	0	0		
C	67,2	70,6		
H	7,9	9,0		
N	3,3	4,3		
Cl	0	0		
S	0,9	0,9		
O	20,7	15,2		
Calorific value (MJ/kg)	33,2	35,2	24,0	34,5

[9, 89]

Table A2: Composition of syngas, assumed the same for digested and undigested sludge due to lack of data.

	300C	**500C**
Chemical composition (wt. %)		
CO_2	100	42
H_2	0	31
CO	0	15
CH_4	0	10
C_2H_4	0	2

[60]

B Measured concentrations of the HOCs and HMs in Norwegian sewage sludge

Table B1: Measured mean concentrations (μg/kg DM) of the selected pollutants in Norwegian sewage sludge in 2017/2018.

HOC	μg/kg DM	Name in USEtox on SimaPro	CAS-number	Impact categories
DEHP	27601	Phtalate, dioctyl-	000117-81-7	HTPc, HTPnc, FETP
PFOS	13,5			HTPc, HTPnc, FETP
PFOA	1,3			HTPc, HTPnc, FETP
SCCP	572			FETP
HHCB	9979			FETP
AHTN	2499			FETP
OTNE	14000			FETP
BDE-209	406	Decabromodiphenyl oxide	001163-19-5	HTPc, HTPnc
PCB7	17	1,1'-Biphenyl, 2, 4-dichloro-, PCB-7	033284-50-3	FETP
NP+NPE	4127	Nonylphenol	025154-52-3	FETP
BPA	1605	Bisphenol A	00080-05-7	HTPc, HTPnc, FETP
PAH16	1660	see table B2		
Triclosan	984	5-Chloro-2-(2,4-dichlorophenoxy)phenol	003380-34-5	FETP
LAS	14911,5	Alkylbenzenesulfonic acid, sodium salt c10-c13	068411-30-3	FETP

[20, 24]

Table B2: Assumed mean concentrations (μg/kg DM) of the individual PAHs in PAH_{16} in Norwegian sewage sludge.

PAH	μg/kg DM	% of total PAH	CAS number	Impact categories
phenanthrene	200,00	12,0 %	85-01-8	HTPc, FETP
pyrene	200,00	12,0 %	000129-00-0	HTPc, HTPnc, FETP
fluoranthene	200,00	12,0 %	00206-44-0	HTPc, HTPnc, FETP
naphthalene	200,00	12,0 %	000091-20-3	HTPc, HTPnc, FETP
fluorene	200,00	12,0 %	000086-73-7	HTPc, HTPnc, FETP
acenaphthene	200,00	12,0 %	000083-32-9	HTPc, HTPnc, FETP
benzo(b)fluoranthene	46,00	2,8 %	00205-99-2	HTPc
benzo(a)anthracene	46,00	2,8 %	000056-55-3	HTPc, FETP
chrysene	46,00	2,8 %	000218-01-9	HTPc
indeno(1,2,3-cd)pyrene	46,00	2,8 %	000193-39-5	HTPc
benzo(a)pyrene	46,00	2,8 %	000050-32-8	HTPc, FETP
anthracene	46,00	2,8 %	000120-12-7	HTPc, FETP
dibenz(a,h)anthracene	46,00	2,8 %	000053-70-3	HTPc, FETP
acenaphthylene	46,00	2,8 %	000208-96-8	HTPc
benzo(k)fluoranthene	46,00	2,8 %	00207-08-9	HTPc
benzo(g,h,i)perylene	46,00	2,8 %	000191-24-2	HTPc

[24]

Table B3: Measured mean concentrations of regulated HMs in Norwegian sludge in 2019.

Heavy metal	Concentration (mg/kg DM)
Pb	13,1
Zn	367,1
Ni	13,7
Cd	0,6
Cu	169,5
Cr	18,4
Hg	0,4

[121]

C HOC removal estimates

Table C1: HOC removal rates during mesophilic AD.

HOC	Reduction in concentration (%)	Reference(s)
NP + NPE	40,04	[35–37]
DEHP	33,5	[35, 38]
BPA	79	[39]
BDE-209	42,5	[40, 41]
PAH	50	[42]
Triclosan	35	[43]
PFOS	0	[44]
PFOA	0	[44]
SCCP	0	-
HHCB	65	[45, 46]
AHTN	65	[45, 46]
OTNE	0	-
PCB7	58	[47]
LAS	0	[48]

Table C2: HOC removal rates during dewatering.

HOC	Removal during dewatering (%)	Reference(s)
NP + NPE	0	-
DEHP	5	[54]
BPA	35	[55]
BDE-209	0	-
PAH	0	-
Triclosan	0	[122]
PFOS	0	[49, 50]
PFOA	0	[49, 50]
SCCP	0	[53]
HHCB	0	-
AHTN	0	-
OTNE	0	-
PCB7	0	-
LAS	0	[123]

D HOC flows per FU in the systems

Table D1: HOC flows (mg) into the processes in scenario 1 and 2: AD+PY500

HOC	AD	Dewatering	Drying	Pyrolysis 500C	Land application
DEHP	27601,00	11942,40	11345,28	11345,28	6126,45
PFOS	13,50	8,06	8,06	8,06	4,35
PFOA	1,30	0,78	0,78	0,78	0,42
SCCP	572,00	341,37	341,37	341,37	0,00
HHCB	9979,00	2084,41	2084,41	2084,41	11,26
AHTN	2499,00	521,99	521,99	521,99	2,82
OTNE	14000,00	2924,32	2924,32	2924,32	15,79
BDE-209	406,00	142,96	142,96	142,96	0,00
PCB7	17,00	7,10	7,10	7,10	0,12
NP+NPE	4127,00	1939,61	1939,61	1939,61	10,47
BPA	1605,00	335,25	217,75	217,75	0,00
PAH16	1660,00	574,60	574,60	574,60	15,51
Triclosan	984,00	634,23	634,23	634,23	0,00
LAS	14911,50	8899,18	8899,18	8899,18	5161,53

Table D2: HOC flows (mg) into the processes in scenario 4: PY500

HOC	Dewatering	Drying	Pyrolysis 500C	Land application
DEHP	27601,00	26220,95	26220,95	8652,91
PFOS	13,50	13,50	13,50	4,46
PFOA	1,30	1,30	1,30	0,43
SCCP	572,00	572,00	572,00	0,00
HHCB	9979,00	9979,00	9979,00	32,93
AHTN	2499,00	2499,00	2499,00	8,25
OTNE	14000,00	14000,00	14000,00	46,20
BDE-209	406,00	406,00	406,00	0,00
PCB7	17,00	17,00	17,00	1,40
NP+NPE	4127,00	4127,00	4127,00	13,62
BPA	1605,00	1042,45	1042,45	0,00
PAH16	1660,00	1660,00	1660,00	27,39
Triclosan	984,00	984,00	984,00	0,00
LAS	14911,50	14911,50	14911,50	8648,67

E Additional inventory calculation parameters

Table E1: Default activity data and from the IPCC methodology for estimating N2O emissions from land application of SS.

Parameter	Value
$Frac_{leach}$	0,3
$Frac_{gasm}$	0,2
EF_1	0,0125
EF_4	0,025
kg N2O-N to kg N2O factor	44/28

[6]

Table E2: Parameters used to estimate the fertilizer replacement with biochar addition to soil.

Parameter	Unit	Value
Initial application rate N [86]	kg/ha	111
Initial application rate P2O5 [86]	kg/ha	48
Initial application rate K2O [86]	kg/ha	96
Application rate of biochar	kg/ha	5000
Assumed reduction in fertilizer supply [85]	%	10
Reduced N2O emissions [87]	kg N2O/kg N fertilizer replaced	0,042

Table E3: Method for estimating the electricity consumption in H2 compression for bio-oil upgrading.

Estimation of electricity for H2 Compression [96]			
Total nominal power in compressors	12276	kW	[96]
Use factor (estimation)	70%	estimated factor	
Power used in H2 compression	8593	kW or kJ/s	[96]
Plant operation	330	days operation	[96]
Plant capacity	2000	dry ton biomass/day	[96]
Bio-oil production from fast pyrolysis	1316	dry ton bio-oil/day	[96]
Bio-oil production from fast pyrolysis	15,23	kg Bio oil/s	[96]
Rate of hydrogen use in upgrading	7,60%	kg H2/kg bio-oil dry	[96]
Hydrogen consumption in upgrading	1,158	kg H2/s	[96]
	7423,34	kJ power/kg H2	[96]
	0,00027778	conversion from kJ to kWh	[96]
Electricity consumption per mass of H2	2,062	kWh/kg H2	[96]

Data gathered by Marcos Watanabe [124].

Table E4: Yields in biochar yield sensitivity analysis in AD+PY500.

Slow pyrolysis at 500°C of digested SS			
Product	Yield wt% DM	-15% char	+15% char
Biochar	54 %	45,90 %	62,10 %
Bio-oil	26 %	30,58 %	21,42 %
Reaction water	12 %	14,11 %	9,89 %
Syngas	8 %	9,41 %	6,59 %

Table E5: Yields in biochar yield sensitivity analysis in AD+PY300.

Slow pyrolysis 300°C of digested SS			
Product	Yield % of DM	-15% char	+15% char
Biochar	75	63,75	86,25
Bio-oil	14	20,3	7,7
CO2	4	5,8	2,2
Water	7	10,15	3,85

Table E6: Yields in biochar yield sensitivity analysis in PY300.

Slow pyrolysis 300°C of undigested SS			
Product	Yield wt% DM	-15% char	+15% char
Biochar	52	44,20	59,80
Bio-oil (dry)	10	11,63	8,38
Water	35	40,69	29,31
CO2	3	3,49	2,51

Table E7: Yields in biochar yield sensitivity analysis in PY500.

Slow pyrolysis 500°C of undigested SS			
Product	Yield wt% DM	-15% char	+15% char
Biochar	33,00	28,05	37,95
Bio-oil (dry)	18,00	19,33	16,67
Water	45,00	48,32	41,68
Syngas	4,00	4,30	3,70

Table E8: Calculation of carbon flows from AD process.

Parameter	Value
% wt. C in CO2	0,273
% wt. C in CH4	0,749
kg CH4/hr in biogas	185,553
kg CO2/hr in biogas	275,325
kg C/hr biogas	214,087
kg C/hr input SS	367,200
kg C/hr in digested SS	153,113

F LCA inventory

Table F1: LCA inventory per FU for all scenarios.

Inputs	**Unit**	**Ref. case**	**Sc 1**	**Sc 2**	**Sc 3**	**Sc 4**	**Sc 5**
Heat	MJ	8857,390	9621,290	9621,290	9342,930	10153,450	9686,750
Electricity	kWh	283,400	283,400	364,520	364,520	30,000	30,000
FeCl3	kg	24,420	24,420	24,420	24,420	25,000	25,000
Polymer	kg	5,140	5,140	5,140	5,140	5,260	5,260
Road transport	tkm	64,890	48,240	48,240	67,140	49,500	52,000
Spreading	kg	648,870	322,270	322,270	0,000	330,000	0,000
NG	Nm3	0	0	9,264	0		0
Outputs	**Unit**	**Ref. case**	**Sc 1**	**Sc 2**	**Sc 3**	**Sc 4**	**Sc 5**
Heat	MJ	5050,440	7999,090	253,720	6437,410	3287,390	1200,000
Electricity	kWh	897,860	1422,070	45,110	1144,440	584,430	213,340
Diesel subst.	kg	0,000	0,000	120,040	0,000	0,000	0,000
Avoided diesel emissions	kg CO2	0,000	0,000	378,6	0,000	0,000	0,000
NG subst.	Nm3	0,000	0,000	238,460	0,000	0,000	0,000
Avoided NG emissions	kg CO2	0,000	0,000	500,33	0,000	0,000	0,000
Coal subst.	kg	0,000	0,000	0,000	268,560	0,000	0,000
CO2 (fossil) from SMR	kg	0,000	0,000	62,998	0,000	0,000	0,000
Negative emissions	kg CO2	0,000	137,200	137,200	283,860	392,400	831,760
N2O emissions	kg	2,367	-0,149	-0,149	0,000	-0,153	0,000
N fertilizer subst.	kg	0,000	0,715	0,715	0,000	0,733	0,000
P2O5 subst.	kg	0,000	0,309	0,309	0,000	0,317	0,000
K2O subst.	kg	0,000	0,619	0,619	0,000	0,634	0,000
CH4 loss to air	kg	5,380	5,380	7,180	5,380	0,000	0,000
CO	g	885,290	1402,161	44,478	1128,418	576,248	210,353
NOx	g	737,143	1167,519	37,035	939,585	479,817	175,152
SO2	g	394,161	624,289	19,803	502,409	256,565	93,656
NMVOC	g	122,109	193,402	6,135	155,644	79,482	29,014
Pb	bioavail. mg	260,249	120,688	120,688	139,577	120,688	139,577
Zn	bioavail. mg	52442,857	24427,399	24427,399	35042,900	24427,399	35042,905
Ni	bioavail. mg	817,459	269,639	269,639	334,190	269,639	334,191
Cd	bioavail. mg	29,290	7,128	7,128	9,345	7,128	9,345
Cu	bioavail. mg	8839,622	143,870	143,870	246,427	143,870	246,427
Cr	bioavail. mg	778,320	187,937	187,937	256,067	187,937	256,067
Hg	bioavail. mg	1,600	0,000	0,000	0,000	0,000	0,000
DEHP	mg	11345,281	6126,452	6126,452	0,000	8652,914	0,000
PFOS	mg	8,057	4,351	4,351	0,000	4,455	0,000
PFOA	mg	0,776	0,419	0,419	0,000	0,429	0,000
SCCP	mg	341,370	0,000	0,000	0,000	0,000	0,000
HHCB	mg	2084,414	11,256	11,256	0,000	32,931	0,000
AHTN	mg	521,991	2,819	2,819	0,000	8,247	0,000
OTNE	mg	2924,320	15,791	15,791	0,000	46,200	0,000
BDE-209	mg	142,957	0,000	0,000	0,000	0,000	0,000
PCB7	mg	7,102	0,959	0,959	0,000	1,403	0,000
NP+NPE	mg	1939,600	10,474	10,474	0,000	13,619	0,000
BPA	mg	217,746	0,000	0,000	0,000	0,000	0,000
PAH16	mg	574,599	15,514	15,514	0,000	27,390	0,000
Triclosan	mg	381,713	0,000	0,000	0,000	0,000	0,000
LAS	mg	8899,183	5161,526	5161,526	0,000	8648,670	0,000

G Input data for estimating characterization factors with the USEtox method

Table G1: Data used to calculate the characterization factors of the three musks HHCB, AHTN and OTNE.

Substance	**Galaxolide (HHCB)**	**Tonalid (AHTN)**	**Iso E Super (OTNE)**
CAS-number	1222-05-5	1506-02-1	54464-57-2
Physico-chemical properties [20]			
MW (g/mol)	258	258	234
Kow	251189	199526	190546
Koc (L/kg)	8710	24547	5981
KH25C (P.m3/mol)	37,18	37,18	0,882034125
Pvap25 (Pa)	0,07266[a]	0,06825[a]	0,19995[a]
Sol25 (mg/L)	1,75	1,25	0,00016
Environmental degradation			
kdegA (s-1)	3,90E-05	2,55E-05	1,94E-04
kdegW (s-1)	5,07E-06	1,23E-06	1,30E-07[c]
kdegSd (s-1)	1,02E-07	1,37E-07[b]	1,44E-08[c]
kdegSl (s-1)	4,80E-08	6,17E-07[b]	6,50E-08[c]
Bioaccumulation factor for fish			
Species (of fish)	BAF (L/kg ww) [125]	BAF (L/kg ww) [126]	
Crucian carp	580	670	-
Smallmouth bass	68,5		-
Largemouth bass	88		-
White perch	177		-
Catfish	194,5		-
Rudd	20	40	-
Tench	510	280	-
Eel	290	400	-
Zebra mussel	620	570	-
average:	*283,1*	*392,0*	*337,6*[c]
Toxicity to aquatic life			
Species:	**Daphnia magna [125]**	**Pseudokirch. subcap. [127]**	
Chronic / Acute:	C	A	-
Test (endpoint)	EC50	EC50	-
Duration	21 days	72 h	-
Concentration (mg/l)	0,293	0,835	-
Acute-to-chronic (mg/l)		0,4175	-
Effect	immobilization		-

[a] Predicted by EPIsuite [128]. [b] Using the experimental data found for KdegW and the division factors 2 and 9 from the USEtox manual [114]. [c] Average of corresponding values for HHCB and AHTN.

(*Continues on next page*)

Substance	**Galaxolide (HHCB)**	**Tonalid (AHTN)**	**Iso E Super (OTNE)**
Species:	**Daphnia magna [125]**	**Pseudokirch. subcap. [126]**	
C or A	A	A	-
Test (endpoint)	EC50	EC50	-
Duration	48 hr	72 hr	-
Concentration (mg/l)	0,282	0,6515	-
Acute-to-chronic (mg/l)	0,141	0,32575	-
Effect	immobilization	biomass	-
Geomean of species	*0,203255996*	*0,368782626*	
Species:	**Acartia tonsa [125]**	**Acartia tonsa [126]**	
C or A	C	C	-
Test (endpoint)	EC50	EC50	-
Duration	6 days	6 days	-
Concentration (mg/l)	0,131	0,072	-
Acute-to-chronic (mg/l)			-
Effect	development	development	-
Species:	**Acartia tonsa [125]**	**Acartia tonsa [126]**	
C or A	C	C	-
Test (endpoint)	EC50	EC50	-
Duration	5 days	5 days	-
Concentration (mg/l)	0,059	0,026	-
Acute-to-chronic (mg/l)			-
Effect	development	development	-
Geomean of species	*0,087914731*	*0,043266615*	
Species:	**Lampsilis cardium [125]**	**Brachydanio rerio [126]**	
C or A	A	A	-
Test (endpoint)	EC50	EC50	-
Duration	48 hr juveline	96 hr	-
Concentration (mg/l)	0,492	0,18	-
Acute-to-chronic (mg/l)	*0,246*	*0,09*	-
Effect	growth	hatching	-
Species:	**Pseudokirch. subcap. [125]**	**Daphnia magna [126]**	
C or A	A	A	-
Test (endpoint)	EC50	EC50	-
Duration	72 hr	48 h	-
Concentration (mg/l)	0,85	0,8	-
Acute-to-chronic (mg/l)	*0,425*	*0,4*	-
Effect	growth		-
Species:	**Paracentrotus lividus [125]**	**Lumbriculus variegatus [126]**	
C or A	A	A	-
Test (endpoint)	EC50	EC50	-
Duration	72 hr	5 days	-
Concentration (mg/l)	0,004	0,397	-
Acute-to-chronic (mg/l)	*0,002*	*0,1985*	-
Effect	development	immobilization	-
average logEC50:	*-1,085508189*	*-0,788602723*	*-0,937055456*

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