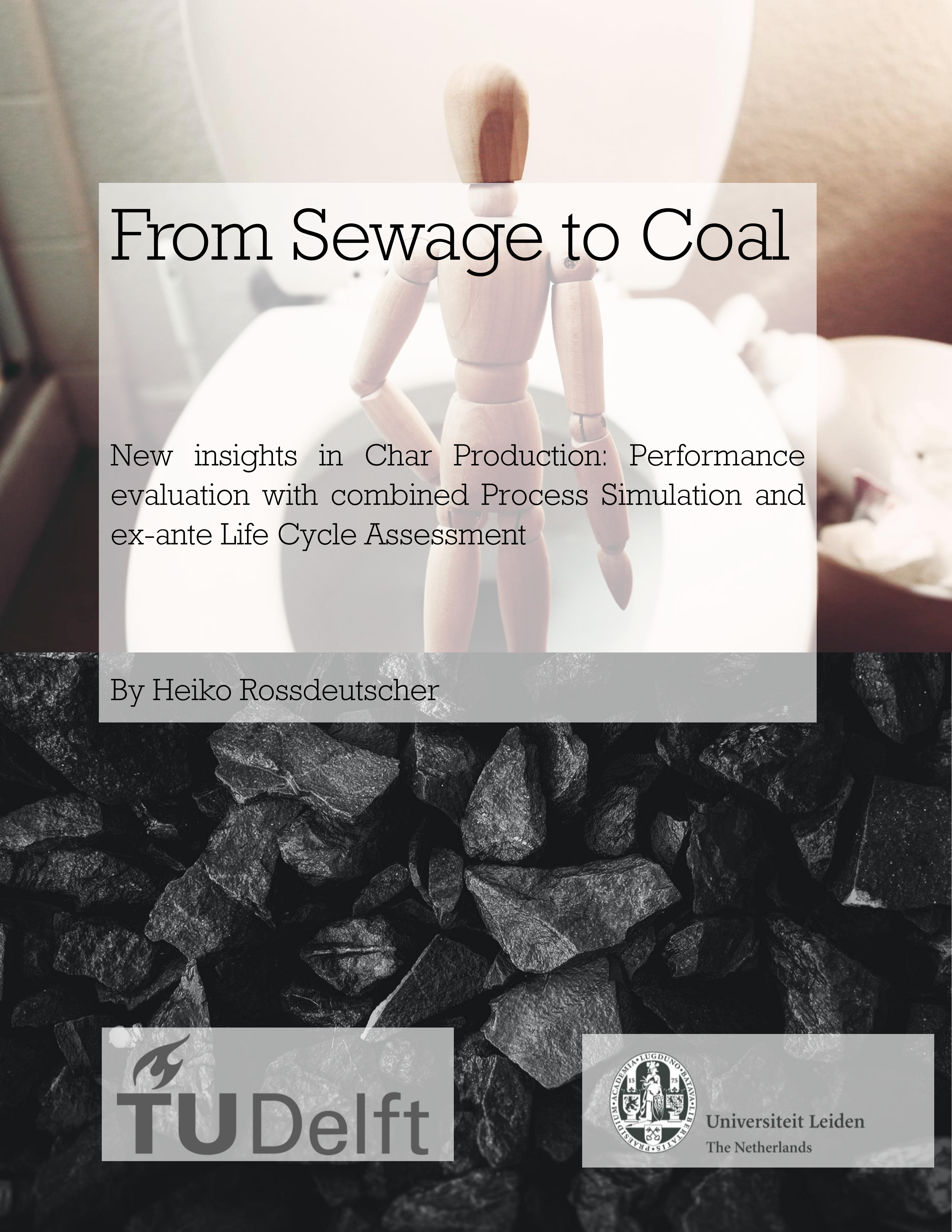

From Sewage to Coal

New insights in Char Production: Performance evaluation with combined Process Simulation and ex-ante Life Cycle Assessment

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Executive summary

The aim of this study was to identify the most sustainable way to produce char from sewage sludge that later can be used as a coal substitute for the steel industry. For this purpose, a literature review was conducted on the latest technological developments for the production of char, which were subsequently simulated in Aspen Plus to obtain process-specific details. The results of the process simulation are then combined with the Life-Cycle-Assessment (LCA) database EcoInvent to assess the environmental impact of the entire value chain for all countries of the European Union using the LCA methodology according to the ISO 14040 framework and the environmental footprint impact family. This family consists of 19 impact categories of which five relevant categories were chosen: (I) climate change, (II) acidification, (III) freshwater ecotoxicity, (IV) freshwater eutrophication and (V) marine eutrophication. In a second analysis the LCA is combined with the integrated assessment model REMIND, which represents the technological evolution in the future. This allows to evaluate the environmental impact in the years 2030 and 2040.

From the literature review, two technology developments that significantly can reduce the overall electricity consumption of the process were identified: an industrial dryer that can dry sewage sludge at 90% efficiency and a heat pump that can convert moist exhaust air into process heat with an efficiency of 400%. Furthermore, the Torwash process was identified, as a pre-processing step for sewage sludge, which increases the efficiency of the filter process and the subsequent drying of the sludge. Based on the results of the literature research, three different technology scenarios were defined. (I) In the baseline scenario, the gases and bio-oils formed during the slow pyrolysis reaction are burned to produce sufficient process heat for the pyrolysis unit and the drying step. (II) In the Torwash scenario, the feedstock is thermally treated before the filtration, which allows a higher solid content in the filter cake and thus reduces the energy consumption in the drying step. (III) The bio-oil scenario considers the condensation of bio-oils and provides the missing process heat with electric heaters.

The process simulation in Aspen Plus was used to determine several process-specific details, including the energy content of the combustion process and the resulting CO_2 emissions. Moreover, the overall energy balance of the different scenario was calculated and the need for external heat was determined. The process simulation additionally revealed a factor not described in the literature yet that influences the energy consumption of the pyrolysis reaction. The higher the ash content of the feedstock, the less energy is required for the pyrolysis process.

In the LCA, the simulation results were combined with further background processes and environmental information from the LCA database EcoInvent and compared against a business-as-usual alternative. In this alternative, the sludge is incinerated and coal with a comparable heating value to the produced char is provided. The results show that the baseline scenario performs better than the business-as-usual alternative in all impact categories. The Torwash scenario, due to the high energy requirements of the pretreatment, performs worse than the baseline scenario in all impact categories and in all but freshwater eutrophication also worse than the business-as-usual alternative. For the bio-oil scenario, the type of heat supply and the CO_2 intensity of the electricity grid is of great importance. If an inefficient resistant heater is used, the bio-oil scenario performs better than the baseline scenario in only half of the European countries, depending on the specific CO_2 intensity of the national electricity mixes. If, instead, a highly efficient heat pump is used, the environmental impact is lower for all EU member states for all impact categories. The difference between the baseline and the bio-oil scenario becomes even larger when the environmental impact is determined for 2030 or 2040. If a conservative integrated assessment model which leads to 2.5°C warming by 2050 is used as the basis for technological change, the emissions of the

bio-oil scenario are 3.020 kg CO_2eq per ton of char, more than 9% lower than the baseline scenario and almost 42% lower than the business-as-usual alternative. In addition, to the ton of char, 120 l of bio-oil are produced, with an economic value that offsets the additional cost of electricity.

Therefore, the strong recommendation is expressed to produce char of sewage sludge according to the bio-oil scenario, with includes the latest drying and heat-pump technologies as well as the co-production of bio-oil.

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1. Introduction

The steel industry plays an important role in global carbon emissions and is responsible for around 7-9% of total emissions (Holappa, 2020). Projections indicate that carbon emissions from the industry will increase significantly due to a projected 4,3% annual growth in global steel demand (Van Ruijven et al., 2016). This increase poses a significant challenge to achieving the global climate targets of the Paris Agreement, which aims to limit global warming to 1,5°C above pre-industrial levels (UNFCCC, 2015). The majority of emissions in the steel industry fall under the Scope 1 and Scope 2 categories. Scope 1 emissions arise from the combustion of coal in high-temperature processes during iron reduction, while Scope 2 emissions refer to indirect emissions resulting from electricity consumption throughout the steel value chain (Teske, Nagrath, et al., 2022). These emissions amount to approximately 1,8 to 1,92 metric tons of CO_2 per metric ton of crude steel, along with other pollutants such as sulfur oxides (SO_x) and nitrogen oxides (NO_x) (Toktarova et al., 2020; Van Ruijven et al., 2016; Worrell et al., 2007). Despite the increasing concern over carbon emissions, steel remains the preferred material for various critical applications such as infrastructure development, mechanical equipment, automobiles, and rail transportation due to its techno-economic advantages (Sambandam et al., 2022; Teske, Niklas, et al., 2022). As the global demand for steel is expected to increase, reducing emissions in the steel industry is critical to achieving climate change goals on a global scale. Therefore, research on various emission reduction strategies in the steel industry is of great importance.

A further electrification of the current steelmaking process is a possible approach. However, it should be noted that the share of CO_2 emissions from electricity is very small compared to the CO_2 emissions from the reduction reaction and melting with coke in the blast furnace (Sambandam et al., 2022). Furthermore, coke plays an important role as a burden carrier in the blast furnace and can only be replaced to a limited extent (Sambandam et al., 2022). Therefore, further electrification alone may not be sufficient to significantly reduce emissions in the steel industry.

Another approach to reduce emissions in the steel industry is to increase the recycling rate of steel. This can be achieved through the use of electric arc furnaces, which can lead to emission reductions of up to 75 % when natural gas is used to generate electricity and close to zero when renewable energy is used (Sambandam et al., 2022). However, the availability of scrap limits the potential for secondary steel production. While the global collection rate for steel scrap is currently 85%, it only meets 35% of steel demand in 2019 and is projected to increase to only 48% by 2050 (Teske, Niklas, et al., 2022). As a result, the majority of steel production still relies on primary production methods, necessitating the exploration of new technologies.

One such technology is hydrogen-based reduction processes in direct reduction iron (DRI) plants. By substituting coal with hydrogen as a reducing agent, a different reaction path can be made possible, with the reduction process taking place in a solid state at a much lower temperature than in conventional blast furnace processes, which can significantly reduce CO_2 emissions (Sambandam et al., 2022).

The demand for hydrogen cannot be met with conventional production methods because they cause high CO_2 emissions. Methane steam reforming and coal gasification are the main methods of hydrogen production, but they release significant amounts of CO_2 (Burmistrz et al., 2016; IEA, 2021) . Carbon capture utilisation and storage (CCUS) technology is available but not yet widely deployed due to high investment costs and energy requirements, uncertain storage capacities and emissions potential (IEA, 2019; UNECE, 2021). Electrolysis is a promising alternative for producing hydrogen without CO_2 emissions, especially if renewable energy sources are used. Although only a small percentage of hydrogen was produced by electrolysis in 2021, the number of projects under development is increasing and is expected to increase further in the coming years (IEA, 2021). However, more hydrogen production methods need to be explored to meet the increasing demand for hydrogen.

One promising method is methane decomposition, a process by which methane is broken down into H_2 and solid carbon. Unlike the SMR technology where the carbon is released in form of gaseous CO_2 and thus contribute to the greenhouse effect, the methane decomposition secrete the carbon in solid form and thus have no contribution to the greenhouse effect. Furthermore, this process is thermodynamically less energy-intensive than the electrolysis of water (Chemistry LibreTexts, 2022), which is an advantage in view of the faltering expansion of renewable energies.

Three different methane decomposition processes are described in the literature: Plasma, thermal and catalytic decomposition. While plasma decomposition requires temperatures of up to 2000°C, thermal decomposition only requires temperatures between 1000°C and 1300°C (Bromberg et al., 1998; Guéret et al., 1997). The presence of a catalyst can further reduce the process temperature to about 900°C and accelerate the reaction (Patel et al., 2020). However, the catalyst used in this process is often quickly deactivated by the solid carbon deposits that block its catalytic capabilities. The regeneration of the catalyst is energy intensive, thus reducing the economic viability. To address these concerns, research into an organic catalyst which does not need to be regenerated, is currently being carried out as part of the H2Steel project (H2Steel, 2022).

The research is focused on creating a cheap organic catalyst from waste streams. One of the waste streams under investigation is sewage sludge, a by-product of wastewater treatment, and in itself a waste stream that poses a serious threat to the environment due to its physio-chemical and sanitary properties (Królik et al., 2019). The current end-of-life options for sewage slud are either landfilling, utilization in agriculture or incineration (Werther & Ogada, 1999), but all of them come along with their specific problems. Landfilling, the most traditional and widespread disposal method in EU-12 countries (new member states that joined EU after 2004) (Kelessidis & Stasinakis, 2012), can lead to secondary pollution of soil and water resources and is therefore no longer allowed in many countries (Park et al., 2008; K. Yang et al., 2017). Agricultural use of sewage sludge is considered one of the basic and most cost-effective methods of management due to its soil-building and fertilising properties. However, the presence of harmful contaminants and microbial pathogens in food raises concerns about the potential risk to human health and the natural environment (Krzyzanowski et al., 2016). While incineration is a mature technology for sewage sludge disposal, it still faces problems such as pollutant emissions and high costs (Liang et al., 2021). Moreover, the European Directive 2018/851/EU establishes a hierarchy for waste prevention and management, giving preference to prevention, preparation for re-use and recycling before other recovery and finally disposal processes (Collivignarelli et al., 2019). This makes the dominant regime, incineration, a not favourable option for future treatment of sewage sludge and opens the door for innovative processes. The novel process of the H2Steel project combines the need of one industry with the use of a waste stream of another as feedstock, thus providing a holistic circular approach (H2Steel, 2022).

1.1. The H2Steel Project

The H2Steel project is a joint project of seven European universities and industrial partners that aims at the sustainable production of green hydrogen and biocoal from circular bio-waste streams. The process uses both biomethane and a catalyst derived from circular bio-waste sources, such as sewage sludge. These are subjected to anaerobic digestion to produce biogas and digestate. While the biogas is purified and fed into the grid, the digestate is further processed to produce a solid residue called char. The char is obtained in a slow pyrolysis process, which breaks down organic compounds into condensable, non-condensable gases and char. The gases are burned to sustain the endothermic reaction, while the char is further refined by chemical leaching to remove elements such as calcium, phosphorus and nitrogen that negatively affect the properties of the catalyst. The extracted elements have considerable value as basic ingredients of fertilisers and can be marketed as such. The upgraded char, which contains a purified organic content, serves as a catalyst for the next process, the methane decomposition process (H2Steel, 2022).

In the biomethane cracking unit developed by the H2Steel project, biomethane is decomposed into hydrogen and solid carbon. The upgraded char serves as an organic catalyst for this reaction and solid carbon is deposited on its surface. As this deposition leads to a deactivation of the catalyst, the catalyst has to be constantly renewed. Unlike metallic catalysts, which require costly removal of carbon deposits, the presence of carbon deposits on organic catalysts is considered beneficial. In fact, carbon deposits can increase the carbon content of the catalyst to as much as 80%, converting the upgraded char into a coal substitute, which for example can be used in the steel industry.

While the entire value chain is being simulated and experimentally tested during the three years of the H2Steel project, this five months master's thesis is devoted only to the first link in the value chain, the production of char. The goal thereby is to find the most sustainable process chain to produce char.

1.2. Char Production via Slow Pyrolysis

The feedstock investigated in this report is sewage sludge, which is digested in biogas plants using anaerobic digestion. In this process, about half of the organic matter in the sewage sludge is converted into biogas while the other half remains in the digested sludge (Jules van Lier, 2023a). While the biogas can be fed to the grid, the digested sludge has a high moisture content and must therefore be dewatered before it can be further processed. The most common technologies for this are centrifugation and chamber filtration. Those mechanical dewatering methods can reduce the moisture content from 96% to 75% and 65%, respectively, transforming the liquid sludge into a solid filter cake (Jules van Lier, 2023a). In order to further reduce the water content, thermal drying is required. The heat required for the evaporation of the remaining water can be extracted from the hot exhaust gases of the pyrolysis process.

Pyrolysis is a process in which organic material is decomposed in an inert atmosphere at elevated temperatures. The organic material is converted into three products: a solid product called char, a condensable vapour product and a non-condensable gas product (Barry et al., 2019; Hagner et al., 2020). The non-condensable gas stream consists mainly of CO, CO_2, CH_4 and H_2 and is normally burnt to generate heat for the pyrolysis reaction (Barry et al., 2019). If further heat is needed the condensable gases can also be burnt. If this is not the case, condensable gases can be refined and converted into bio-oil or special chemicals. The solid char consists mainly of carbon and the inorganic components (ash) of the biomass (Bridgwater, 2012).

Depending on the operation conditions, the pyrolysis process can be classified in either fast pyrolysis, temperatures around 500°C and short hot vapor residence time (~1 s) or slow pyrolysis, which has typically lower operating temperatures (290 – 400°C) and a long vapor residence time (>10 min) (Bridgwater, 2012). The two processes focus on different products: Fast pyrolysis produces more condensable gases, while slow pyrolysis produces more char (Bridgwater, 2012). As the H2Steel project aims to produce char, slow pyrolysis technology is used.

The energy obtained by the burning of the pyrolysis gases is partly feed back into the process to sustain the endothermic pyrolysis and drying process. The other part of the hot gases is passed through heat exchangers gas and purification steps to extract the extra heat and clean the exhaust gases from sulphur and nitrogen oxides as well as other harmful pollutants, before it is emitted to the air. The extra heat can be used by other processes in the H2Steel value chain.

In order to find the most sustainable way of char production, a literature review was conducted to explore the state of the art and identify the knowledge gap.

1.3. State of the Art

Several thermal conversion processes are being studied for the use of biomass, but pyrolysis has prevailed due to the good quality of the end products it produces (Ward et al., 2014). To determine the areas of application for the pyrolysis process, various biomass feedstocks have been investigated, ranging from organic wastes in the paper industry, to various agricultural waste streams and sewage sludge (Barry et al., 2019; Carrasco et al., 2017; J. Han et al., 2017a, 2017b; Lan et al., 2018). These feedstocks differ in composition and initial moisture content, resulting in different pre-processing steps and different ratio of the pyrolysis products.

The interaction between reaction temperature, pressure and time has also been extensively researched in previous studies (Barry et al., 2019; Bridgwater, 2012; Trinh et al., 2013). However, due to the higher economic value of biofuel compared to biochar (Yoder et al., 2011), most of these studies have focused on higher operating temperatures to increase the biofuel yield (Bridgwater, 2012). Moreover, most of those studies focus on technical and economic aspects of the technology rather than the environmental impact of it. Next to process simulations carried out in the simulation software Aspen Plus, there are also experimental approaches being explored. Park (2008), for example, investigated 12 different operating conditions for the feedstock sewage sludge, gaining new insights into product distribution over different operating conditions.

There are not many scientific studies on slow pyrolysis at temperatures below 400°C. However, the existing studies provide valuable information on how the relationship between char, bio-oil and gases behaves at temperatures between 300 and 500°C (Barry et al., 2019). These show that lower temperatures and longer process times increase the char yield. In addition, Park (2009) describes the composition of charcoal at different temperatures, suggesting that lower temperatures lead to a higher carbon content in the char. Since the final purpose of char is to be a reducing agent for the steel industry, a high carbon content is desirable.

To determine the environmental impact of a technology, life cycle assessment (LCA), which takes into account all upstream and downstream emissions, is common practice (European Commission, 2015). Several studies can be found in the literature that assess the environmental performance of the utilisation of different biomass feedstocks (Mogensen et al., 2014; Parajuli, Knudsen, et al., 2017; Parajuli, Kristensen, et al., 2017). The uses range from electricity generation(Badri et al., 2012; Fan et al., 2011; Giuntoli et al., 2016; Sastre et al., 2014) to biogas (Boschiero et al., 2016; Cherubini & Ulgiati, 2010; Wang et al., 2016; J. Yang & Chen, 2014) and biofuel production (Abbas & Handler, 2018; Dang et al., 2014; Peters et al., 2015b; Schmidt, 2015). These products are compared against the production of electricity via a combustion process, the production of natural gas or petroleum fuel respectively. Only a few studies combine a process simulation with an LCA to compare the production of char to the ordinary combustion of sewage sludge (Barry et al., 2019; Hospido et al., 2005).

1.4. Knowledge Gap

As far as the author is aware, there are no reports in which different LCA scenarios are used to determine the most environmentally sustainable production of char from sewage sludge. Although the technical operation conditions of pyrolysis process itself are described in detail in the literature, there is not sufficient information about the integration of slow pyrolysis into different realistic process chains. While recent technical developments of each technology are described in separate reports and company announcements, there are no collected works summarizing recent developments. To address this knowledge gap, the following main and sub-research questions were formulated:

What is the most environmentally sustainable scenario to produce char from sewage sludge, so that it can be used in the H2Steel process?

- *What are possible upscaled technology scenarios to produce char from sewage sludge?*
- *What process information can be obtained from a process simulation in Aspen Plus?*
- *What is the life cycle environmental impact of the different technology scenarios considering different electricity mixes in various European countries?*
- *What is the life cycle environmental impact of the different technology scenarios in 2030 and 2040, when the ongoing transition of electricity production is considered?*

2. Concept of analysed technologies

In order to form an understanding of the process, interviews were conducted with the Italian partners of the H2Steel project and with wastewater specialists from the TU Delft. A subsequent literature review provided the necessary process details. These are briefly described below for each technology before the scenarios necessary for the subsequent simulations are formed.

2.1. Anaerobic Digestion

Wastewater treatment plants comprise a number of processes for purifying wastewater. A detailed description of the entire wastewater treatment process can be found on the website of the wastewater treatment plant constructor (Hawk, 2023). However, due to the scope of this report, only the last step of the process, the stabilization of the sludge via anaerobic digestion, will be discussed. In this process, the sludge consisting of microorganisms is decomposed in the absence of oxygen. This process releases biogases that can be captured and fed into the natural gas grid as well as residues known as digested sludge. This digested sludge is the feedstock for the technology developed in the H2Steel project. The composition and particle size of the digested sludge influence the subsequent processes. The composition of the digested sludge depends strongly on the industries discharging their waste water into the municipal wastewater treatment plant, which can lead to large regional differences. In order to clearly determine the composition of the sludge, two different analyses are necessary: proximate and ultimate analysis (Dahawi et al., 2019). In the proximate analysis, moisture content (MC), volatile matter (VM), fixed carbon (FC) and inorganic materials summarised as ash (ASH) are determined according to equation 1.

$$MC + VM + FC + ASH = 100\% \qquad [1]$$

In the following report, the moisture content of the sludge is also referred to as the solids content (SC). The relationship between moisture and solid content is described in equation 2.

$$MC + SC = 100\% \qquad [2]$$

For the ultimate analysis, the basic elements of the compounds are examined (equation 3). Where C is the carbon content, H is the hydrogen content, O is the oxygen content, S is the sulphur content and Cl is the choline content.

$$C + H + O + S + CL + ASH = 100\% \qquad [3]$$

The particle size distribution is important to determine the mesh size of the subsequent filtration step, but as with the composition, this varies from region to region. In this report, the digested sludge of the Michigan wastewater treatment plant is used due to the good data basis. The results of the proximate and ultimate analysis as well as the particle size distribution can be found in the Appendix 7.1.1.

2.2. Dewatering Technologies

Immediately after anaerobic digestion, the digested sludge has a moisture content of 96% (EPA, 1998). To prepare the digested sludge for the slow pyrolysis process, the moisture content has to be reduced to less than 10%. For an energy efficient removal of the water, first a mechanical dewatering takes place before the remaining water is evaporated in a drying step.

Mechanical dewatering is only capable of squeezing a certain percentage of water out of the sludge. This is due to the interaction between the sludge particles and the water. These interactions fall into three categories: (I) free water, (II) interstitial water, and (III) surface water. In the case of free water, there are no interactions between the particles and the water, so it can be completely removed by mechanical dewatering. However, the interstitial water, which is within the flocs but not associated with the solids, and the surface water, which is bound to the sludge surfaces by hydrogen bonds, cannot be removed by mechanical dewatering steps (Sludge Processing, 2020). The proportion of free water can be increased by adding flocculants to the digested sludge. These promote the agglomeration of larger flocs and thus reduce the proportion of interstitial water (Kreuzinger, 2020).

For the removal of the water, a filter press or a centrifuge are often used (Jules van Lier, 2023b). Centrifugal dewatering is a continuous process that was developed in the 1930ties. It uses the force from high-speed rotation to separate liquid from the sludge (States Environmental Protection Agency - Office of Water, 2000a). The conical shape of the centrifuge helps to lift solids out of the liquid so that they can dry before being discharged (Kemp, 1997). With this technology, a solid content of 25% can be achieved. This requires 10 kg of flocculant and 100 kWh of electricity per ton of sludge (TU Delft OCW, 2023).

In a chamber filter press, two plates create a chamber to pressurize solids and squeeze out liquid through a filter cloth lining the chamber. After the required moisture content is reached, the press opens, the filter cake falls out of the chambers and the press is ready for another batch. Depending on the required capacity, several plates are suspended from a frame face to face to create a series of chamber (States Environmental Protection Agency - Office of Water, 2000b). Chamber filter presses can achieve a solid content of up to 35%, while requiring only 5 kg of flocculants and 80 kWh of electricity per ton of sludge (Kemp, 1997).

The wastewater from the filtration process has already passed through the first two treatment stages of the wastewater treatment plant. In smaller plants with only 2 treatment stages, this would mean that this water would be discharged directly into the next water carrier. In larger wastewater plants, however, it could go through further processes such as activated carbon filtration, ion exchange or reverse osmosis to further increase the purity of the wastewater (EPA, 1998).

2.3. Torwash

In addition to the established dewatering technologies, research is also taking place with the aim of further increasing the solids content to 50%. One of the promising new technologies is Torwash, a hydrothermal carbonization (HTC) process being developed by TNO. In this process, wet low-grade biomass feedstocks are pressurized and heated up to about 200°C (Van der Meijden & PELS, 2021). This causes the cells of the organic matter to burst, so that subsequent mechanical presses can squeeze not only the free water between the cells, but also the interstitial water. As a result, solids contents of up to 50% can be achieved. In addition, the subsequent filter cake has a higher porosity, pore volume and surface area and a higher calorific value (Zijlstra et al., 2022).

The decisive variables for the reaction are the reaction temperature and the residence time. The higher the reaction temperatures, the more gases (70-90% CO_2) and liquid carbon compounds are produced. Longer residence times, on the other hand, increase the yield of hydrochar, the solid that can be obtained after mechanical dewatering (Heidari et al., 2019; Nizamuddin et al., 2017).

Although the pilot plants described in the literature only have a throughput of 50 kg/h, the results are promising and larger plants are being planned. A prefiltration reduces the moisture content of the feedstock from 96% to 80%. The thick sludge is than passed through 3 sections, a preheating section, a process section and a cooling section. In the preheating section, the feedstock is slowly preheated to 180°C for 30 minutes. In the subsequent process section, the feedstock is heated to 200°C and hydrothermal carbonization reactions take place under autogenous pressure for 1 hour. In the cooling section, the mixture is cooled down to 40°C to stop the reaction (Zijlstra et al., 2022).

2.4. Drying process

To reduce the moisture content from the Torwash process (50%) or the filter press (65%) to below 10%, thermal drying of the filter cake is necessary. In this process, filter cake is heated up to evaporate the remaining water. The temperature at which the drying takes place not only influences the speed of the process, but also the emissions produced. The higher the temperature, the faster the drying process, but also the more decomposition reactions take place in the sludge. These lead to the formation and evaporation of nitrogen, sulphur and organic carbon compounds (Ding et al., 2015). Together they account for approx. 0,3% of the total feedstock.

While conventional dryers for sewage sludge evaporate the remaining water with an efficiency of 75% (Cheng et al., 2020), the company Huber was able to develop a 2 chamber belt dryer with an efficiency of 90% (Huber SE, 2023).

2.5. Slow Pyrolysis

The dried filter cake is transferred directly to the slow pyrolysis reactor. In this reactor, the feedstock is heated up under oxygen exclusion. Thereby, several complex conversion processes take place simultaneously, finally resulting in 3 different products: (I) volatile pyrolysis gases, (II) volatile bio-oils and (III) solid char. The difference between pyrolysis gases, which will be referred to as gases in this report, and bio-oils is their boiling point. If the substances are liquid at room temperature, they are classified as bio-oils (Collard et al., 2016). The operating conditions temperature, heating rate, pressure, and particle size have a significant influence on the ratio of the pyrolysis products.

2.5.1. Char

The solid residue from the pyrolysis reaction is also called charcoal, biochar or activated carbon. It consists mainly of benzene rings in a polycyclic structure that have a high carbon content. In addition to carbon, charcoal also contains the inorganic substances of the source material. The inorganic substances consist mainly of metals and heavy metals. They are considered as ash in this report. The possible uses range from use as a heat source for households and agricultural applications to replacing coal in the steel industry, which is the aim of the H2Steel project.

2.5.2. Bio-oils

In addition to the desired bio-oils, the condensable fraction of the pyrolysis gases consists of up to 80% water. For the use of bio-oils, phase separation must take place, which separates the water from the complex mixture of hundreds of organic compounds (Kanaujia et al., 2014). Many of the organic compounds contain between one and four carbon atoms to which various functional groups are attached (alcohol, ketones, aldehydes or carboxylic acids)(Collard et al., 2016). Bio-oil can be used in many static applications such as furnaces, boilers, engines, or turbines to generate heat or electricity (Meier et al., 2013). Its high oxygen content and varying viscosity, density, acidity, and instability make it however unsuitable as an alternative to conventional fossil fuels in mobile applications (Collard et al., 2016), unless it is upgraded.

2.5.3. Gases

The non-condensable fraction of the volatile pyrolysis products consists mainly of CO_2, CO, CH_4 and H_2. Other short carbon chains such as ethane, ethylene, propane and butane have been reported, but only in small amounts (Neves et al., 2011). All compounds except CO_2 have fuel properties, so this gas fraction is often used to generate part of the energy needed for the pyrolysis process (Collard et al., 2016).

2.5.4. Temperature

Collard et al. (2016) describes in his work detailed the main steps of the biomass conversion when exposed to elevated temperatures under oxygen exclusion. In this work it is also described that at higher temperatures more secondary and tertiary reactions take place which leads to more volatile pyrolysis products. The theoretical work of Collard is also proven in several experiments; for example, Barry et al. (2019) showed a tendency for the oil and gas fraction to increase and the char fraction to decrease at higher temperatures (Figure 1).

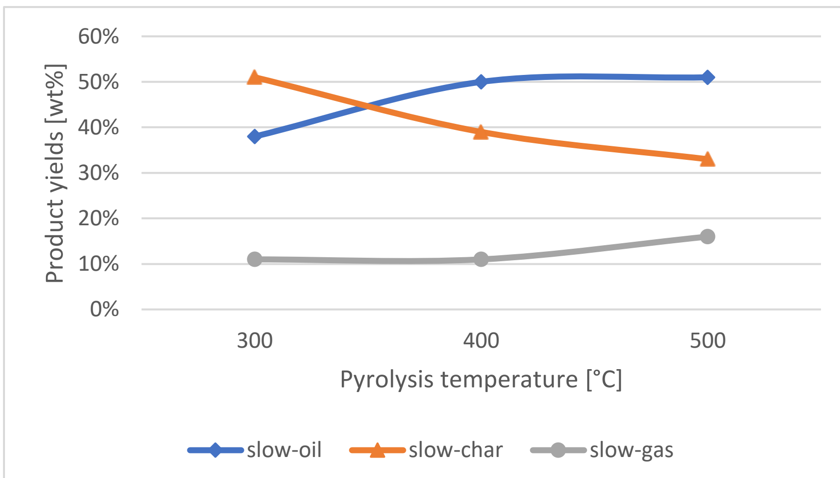

Figure 1 Production yield of slow pyrolysis at different temperatures (remodeled from Barry et al., (2019))

2.5.5. Heating Rate

The biomass feedstock consists of a wide variety of molecules whose chemical bonds vary in strength. At slow heating rates, the weakest chemical bonds break first, and subsequent rearrangement reactions lead to only minor changes in the structure of the polymers. In contrast, at very fast heating rates, many chemical bonds break at the same time. This results in a higher proportion of gases and oils compared to slower heating rates, which produce more char (Collard et al., 2016).

2.5.6. Particle Size

With larger particles, heat and mass transfer occurs more slowly, limiting the heating rate of the particle core. As already described, the slower heating rate results in more char being produced. This is mainly at the expense of the oily products, as the longer material transfer times also allow more secondary and tertiary reactions that promote gas formation (Collard et al., 2016). This trend is also confirmed in the experiments of Park et al. (2008) which results are displayed in Figure 2.

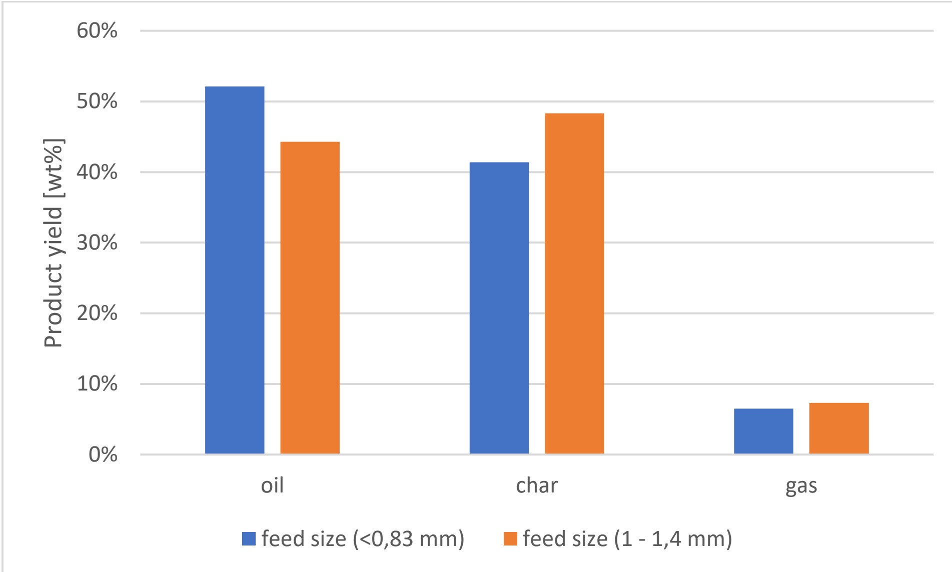

Figure 2 Product distribution at 450°C with different feed sizes (remodeled from Park et al., (2008))

2.5.7. Pressure

To investigate the influence of different pressures on the production of char, Dahawi et al. (2019) carried out a simulation with Aspen Plus. In the simulation, only the pressure in the slow pyrolysis reactor was changed, while all other reaction parameters remained the same. The results showed a slight increase in char yield with increasing pressure. However, Dahawi concludes that this minimal increase does not justify the higher process requirements and energy demand associated with the higher pressure. The higher char yield in Dahawi's simulation results is due to the fact that biomass from empty fruit bunches of oil palm which has a higher initial carbon content was used instead of sewage sludge.

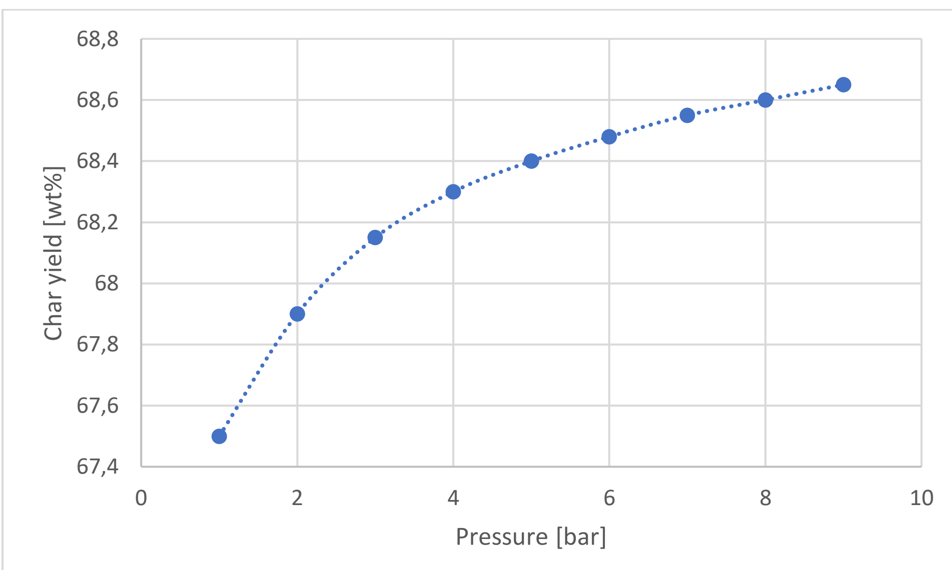

Figure 3 Char yield as a function of pyrolysis pressure (remodeled from Dahawi et al., (2019))

2.5.8. Pyrolysis reactor

The described pyrolysis process can be carried out in different reactors. Typically, the reactors for slow pyrolysis have a bed density close to the density of the feedstock (Collard et al., 2016). This means that a majority of the char produced can be taken directly from the reactor and only the fraction of small and light weight particles is carried on in the gas stream.

2.6. Cyclone

To separate the smaller char particles carried by the gas stream, a cyclone is often used in the industry. A cyclone uses centrifugal forces to push the particles against the cylindrical outer walls of the cyclone. In case of low separation requirements, the upward movement of the hot exhaust gases is often sufficient to generate enough centrifugal force to separate the solid particles from the gas stream without the need for an extra compressor (EMIS, 2022).

Depending on the utilization of the bio-oils, the purified gas stream is either directly directed into the combustion chamber, or previously into a condenser in which the bio-oils are separated from the non-condensable gases.

2.7. Combustion Chamber

In case all volatile components, i.e. gases and bio-oils, are being burnt, the gas flow from the cyclone can directly be directed into the combustion chamber. To achieve complete combustion of the organic compounds, sufficient oxygen must be added to the reaction. In industry, sensors can be used to analyze the composition of the exhaust gases and thus control the amount of fresh air. The fresh air is supplied to the process via a compressor at a slight overpressure. Under these reaction conditions the volatile organic compounds burn at temperatures of more than 1000°C. The smallest char particles that have not been separated from the gas stream in the cyclone also burn at these temperatures and therefore pose no danger to the environment.

In order to optimally utilize the thermal energy contained in the exhaust gas stream, it is passed through several boilers to allow the production of different steam classes.

2.8. Condensation of bio-oils

Instead of combusting both the gases and bio-oils it is also possible to condensate the bio-oils and thus create another product with economic value. Therefore, the gas stream from the pyrolysis reactor flows at 400°C into the condenser. In order to separate the condensable bio-oils from the non-condensable gases, the gas flow is quenched with water. The water cools the gas flow and substances with a higher boiling point condense. A decisive factor for the efficiency of the separation is the contact area between the water and the gas. One way to increase this contact area is to arrange several stages in a column. In addition to the contact area, the temperature of the quench water also plays a significant role. If the temperature is too low, substances such as methane or carbon monoxide are condensed in significant quantities. Since their boiling point is relatively low, they tend to outgas and are therefore not desired in the bio-oil. In order to keep their concentration as low as possible, the quench water is usually preheated to 60°C (Ibarra-Gonzalez & Rong, 2019).

The separation of the bio-oils from the water can take place in several ways. The gravitation method, which does not require energy, makes use of the density differences between oil and water. In a retention tank, the lighter bio-oils rise while the heavier water molecules sink to the bottom of the tank. In this way, an almost perfect separation between the bio-oils and the water can be achieved (Kimray, 2023).

The non-condensed gases are passed on to the combustion chamber and burned there.

2.9. Electric heater

In the case that the bio-oils are condensed and not enough process heat is generated in the combustion chamber, additional heat energy can be provided by an electric heater. Due to the future orientation of this work, the widely used natural gas heaters are not considered, but only electric heaters. In the case of electric heaters, there are great differences in the efficiency with which electrical energy is converted into thermal energy. While widely used resistant heaters convert almost 100% of the electrical energy into thermal energy (NewAir, 2019), newly developed heat pumps can achieve efficiencies of up to 400%. An industrial heat pump, which can reach temperatures of up to 150°C, was developed in the EU project DryFiciency. In this project, the Austrian Institute of Technology and Sintef Energy Research developed a heat pump that can extract energy from moist exhaust air with a residual heat of 70°C and heat dry air to up to 160°C (Bantle, 2020).

2.10. Exhaust gas cleaning

The Europe-wide legal framework for waste-to-energy (BREW WI) requires low emission levels from waste treatment plants. To achieve those emission levels, pollutants have to be removed from the exhaust air. Therefore, exhaust gas cleaning systems, also called scrubbers, are necessary. These systems fall into two main categories: Dry scrubbers and wet scrubbers.

Dry scrubbers do not use liquid substances for the scrubbing process. Instead, hydrated lime granules are used, which are exposed to the flue gas to trigger a chemical reaction that removes sulfur oxides (SO_x) and hydrogen chloride (HCl) with an efficiency of up to 99 %. However, nitrogen oxides, which are produced during combustion processes, cannot be removed with this method.

To remove the nitrogen oxides from the combustion process, a wet scrubbing process with ammonia is required. In this process, the ammonia is diluted with water and sprayed into the gas stream in fine droplets. In large scale combustion plants, hydrochloric acid, iron (III) chloride and sodium hydroxide are also mixed into the water for the wet scrubber to filter other harmful pollutants from the flue gases.

2.11. Development of technology scenarios

Based on the process details obtained, the following three technology scenarios were formed (Figure 4). The baseline scenario uses a chamber filter press to reduces the moisture content of the sludge from 96% to 65%. The filter cake is then dried in a 2-chamber filter belt dryer before being directed to the slow pyrolysis reactor. There, the organic matter is decomposed at 400°C and the char formed can be taken directly from the reactor. Smaller char particles carried by the gas stream are separated from it in a cyclone. Subsequently, both the gases and the bio-oils are combusted to generate the heat required for the slow pyrolysis and drying process.

The Torwash scenario, is based on the baseline scenario, however, the filtration only produces a thick sludge which is than subjected to the thermal treatment. This allows a second filter press to out squeeze more water and thus reduce the moisture content in the filter cake to up to 50%. The extra heat required for the pre-treatment comes from an electric heater.

The difference between the bio-oil and the baseline scenario consists of the condensation of the bio-oils. While the bio-oils are sold as a co-product, the missing heat energy is again provided by an electric heater.

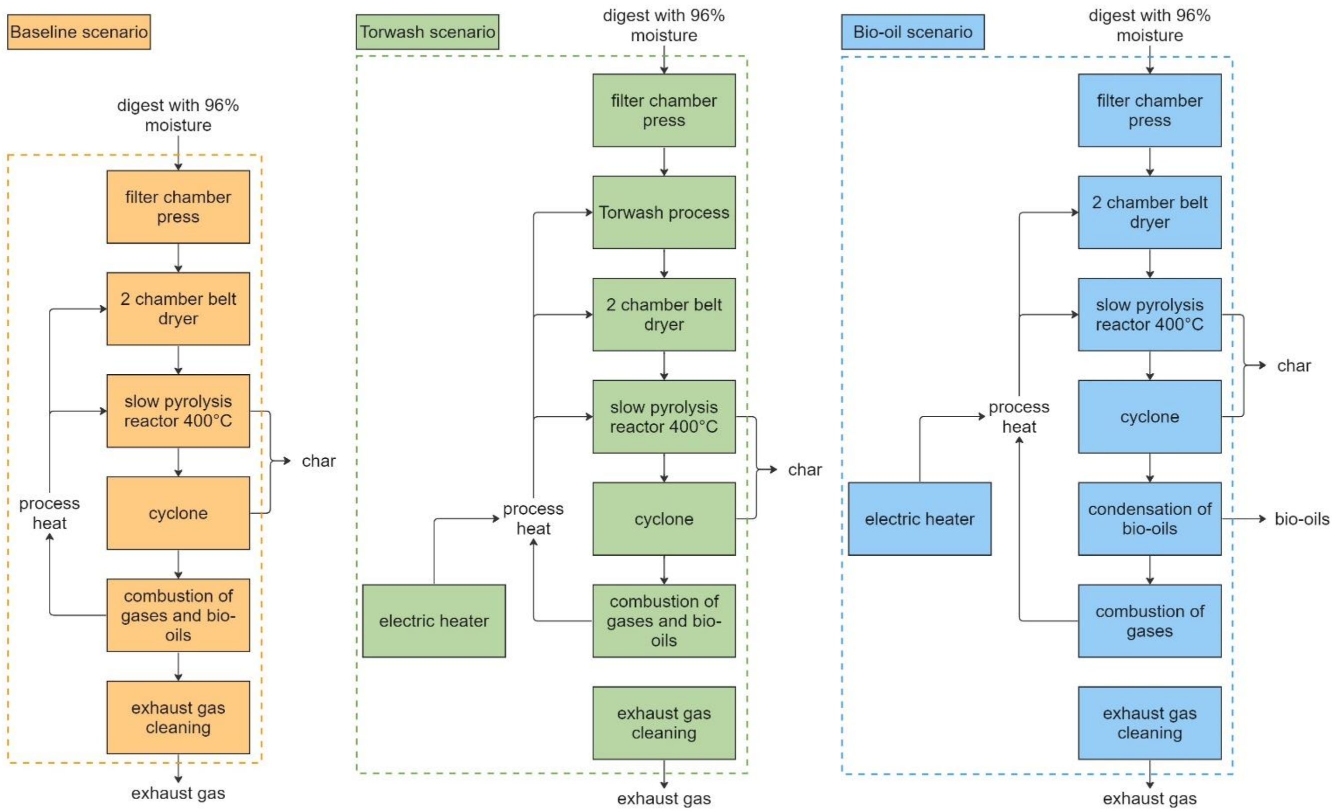

Figure 4 Overview of the technological scenarios

3. Methodology

For a better understanding of the different elements of which this work is composed, a methodological overview is given in Figure 5. The phases shown in the visual representation are related to the sub research questions and are explained in more detail below.

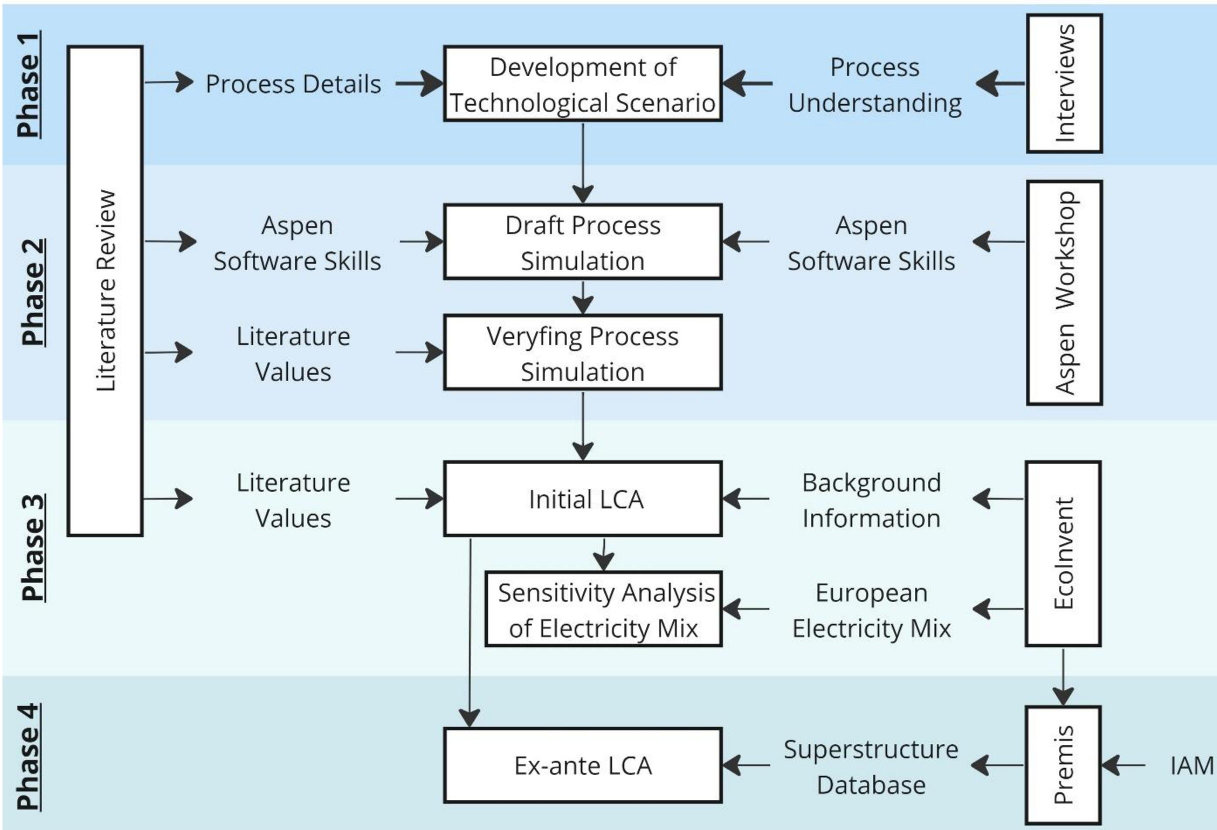

Figure 5 Methodological overview

3.1. Phase 1 – Development of Technology Scenarios

The goal of the first phase was to develop a holistic understanding of the whole process chain from which different technology scenarios can be formed. For this purpose, interviews were conducted with the project managers of the H2Steel project and experts for the treatment of wastewater. The process understanding gained from the interviews was further developed through a literature review. In this review, the necessary process conditions for the subsequent process simulation were also collected.

3.2. Phase 2 - Process Simulation in Aspen Plus

The aim of the process simulation is to combine the different technologies in a process chain and to determine several process-specific details, including the energy content of the combustion process and the resulting CO_2 emissions, as well as the overall energy balance of the different technology scenarios. Due to its comprehensive databases and ability to handle unconventional components such as digested sewage sludge (D. Han et al., 2019), Aspen Plus was chosen as the simulation software. Aspen Plus is a software programme that, thanks to its powerful algorithms and extensive database, can create various biomass models for calculating mass and energy balances of plants and for analysing and optimising process performance (J. Han et al., 2017a). In order to develop the software skills necessary for Aspen Plus, a workshop from Mrs. Gonzalez was attended and literature in which similar Aspen models were described was reviewed. The results of the process simulation are then used to determine the environmental impact.

3.3. Phase 3 - Life Cycle Assessment

For an environmental assessment of the whole value chain of the char production the LCA method is used. This method looks at a variety of environmental impacts, including greenhouse gas emissions, acidification, freshwater ecotoxicity and the eutrophication of fresh and marine water. The holistic approach that includes all life-cycle-stages helps to find ways to minimize these impacts (Guinée, 2002). Performing an LCA on a technology that is still in development brings challenges such as data availability, but also offers the possibility of greater design freedom. This proactive approach is also called ex-ante LCA, or prospective LCA, which is a subcategory of ex-ante LCA and deals with future technologies and their impact (Cucurachi et al., 2018). In order to compensate for the low information availability, prospective LCA include several scenarios to examine the technology. Due to the good possibilities for developing scenarios, the Activity Browser, an open source LCA software based on Brightway2, was chosen to conduct the LCA. To follow the ISO framework, the study reports on the 4 main parts of the framework: goal and scope definition, inventory analysis, impact assessment and interpretation.

3.3.1. Goal and Scope Definition

The goal of the initial LCA is to find the most environmentally friendly functional design for the slow pyrolysis process. The results of this analysis are intended to provide process developers with more background knowledge on the environmental impacts of the different technologies. The geographical scope has been set to Italy, as the experimental work and the first pilot plant will be built there. Most of the input data comes from Aspen process simulation. Since these correspond to a thermodynamic ideal, they do not have a time component, which could be expressed in terms of increased efficiency of a technology. Data on background processes such as treatment of filter water or electricity production are retrieved from the EcoInvent 3.8 database and represent the technological status as of September 2021.

The technology analyzed in this LCA has two functions: (I) the end-of-life treatment of digested sewage sludge, and (II) the provision of a coal alternative for the steel industry. The functional unit has been defined as producing 1000 kg of char with a calorific value of 7,5 MJ/kg HHV from digested sewage sludge with a 96% moisture content. The Torwash and Bio-Oil scenarios use the same input and produce a char with the same characteristics as the baseline scenario. If extra heat is required, an electric heater is used. The business-as-usual scenario also uses the same input, but then burns the filter cake with a moisture content of 73%. To fulfill the second function, the provision of a coal alternative, the business-as-usual alternative provides coal with a comparable heating value to 1000 kg char. Below the most important characteristics of each scenario are displayed:

- **Baseline scenario**: producing 1000 kg char and burning both the pyrolysis gases and bio-oils.
- **Torwash scenario**: producing 1000 kg char with the Torwash preprocessing step and burning both the pyrolysis gases and bio-oils.
- **Bio-Oil scenario**: producing 1000 kg char while condensing the bio-oils and burning the pyrolysis gases.
- **Business-as-usual alternative**: incineration of the digested sewage sludge and provision of coal

3.3.2. Multifunctionality and allocation

Slow pyrolysis is a multifunctional process as it produces three products: non-condensable gases, bio-oils and char. If both the non-condensable gases and the bio-oils are burned to generate the process heat required for drying and slow pyrolysis, they remain in the system and thus do not represent a multifunctionality that needs to be resolved. However, this changes when the bio-oils are condensed and sold as a co-product. In this case, the system under investigation produces two products with economic value. To solve this multifunctionality, the Iso standard 14044 offers a hierarchy of different approaches (ISO, 2006):

1. **Subdivision into smaller processes** was not possible as char and bio-oil are produced in the same process.
2. **System expansion** was the applied strategy. In case the bio-oil is condensed, the business-as-usual alternative must also perform this function and provide a fuel alternative.
3. **Substitution** was not applied, as a solution with higher hierarchy was found.
4. **Allocation** was not applied, as a solution with higher hierarchy was found.

To cope with the system expansion, an additional alternative was created that also fulfills the third function, namely the provision of oil with a comparable calorific value to the condensed bio-oils.

- **Business-as-usual bio-oil alternative**: incineration of the digested sewage sludge and provision of coal as well as oil

3.3.3. Sensitivity analysis

As the H2Steel project is funded by the EU, it is likely that implementation will be Europe-wide, if successfully completed. Country-specific differences, such as the electricity mix, can have a significant influence on the choice of technology. For this reason, the sensitivity analysis aims to show which technology has the lowest environmental impact in the respective EU member states. It is assumed that the decision whether to produce bio-oils as a by-product has the greatest impact on the electricity demand of the process and hence causes the biggest regional differences. Therefore, the sensitivity analysis compares the best scenarios for the combustion of all volatiles with the co-production of bio-oils at the European level.

3.4. Phase 4 – Ex-ante LCA

In addition to country-specific differences, there are also temporal differences that can influence the technology choice. The influence of time plays a particularly important role in industries that are currently in a state of transition, such as electricity production. Its transformation towards carbon neutrality could strongly influence the decision which technology should be used for the H2Steel project. Especially as the H2Steel process will only become marketable in the next few years and then have a lifespan of more than 20 years.

To forecast this technological change, the Potsdam Institute for Climate Impact Research (PIK) has developed the integrated assessment model REMIND. This numeric model depicts the future development of world economies with a special focus on the development of the energy sector and the impact on our global climate. This includes several scenarios, such as the SSP2-NDC, which reflects current nationally determined contributions leading to 2.5°C warming by 2050. A more positive scenario, which is in line with the Paris Agreement and leads to 1.5°C warming by 2050, is the SSP2-PkBudg500 scenario (PIK, 2023).

Each future scenario leads to changes both in the modeled foreground processes and in the background processes that are retrieved from the LCA database EcoInvent. This results in a new Life-Cycle-Inventory (LCI) database for each future scenario and each time horizon considered. To avoid working with multiple databases Steubing & König developed the superstructure approach. In this methodology, all unique exchanges (elementary and intermediate flows) and processes that exist in all scenario LCI databases are collected and combined in one superstructure database. An associated scenario difference file stores all differences between the scenarios and can be used to transform superstructure database into a database for a specific future scenario (Steubing & de Koning, 2021). For a practical implementation of the methodology the software package Premise was used (Sacchi et al., 2023).

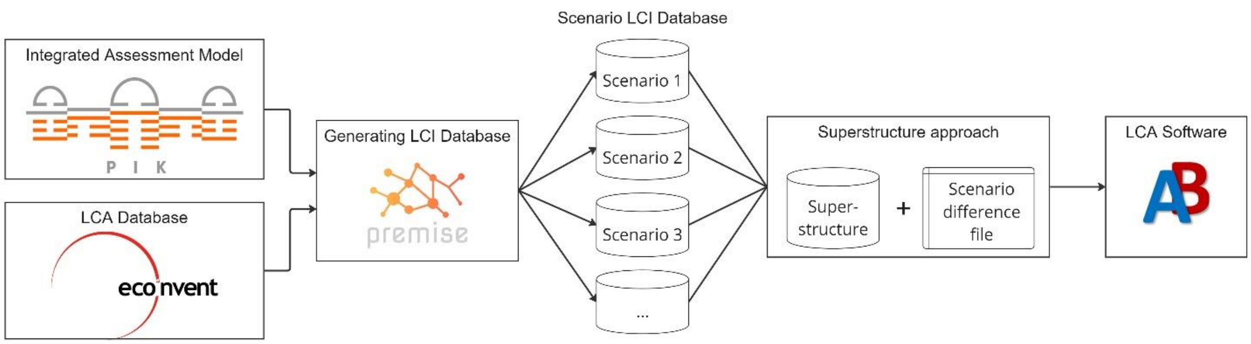

Figure 6 Graphical representation of the generation of the future LCA database

4. Results

In the following chapter, the results of the process simulation are shown and compared with literature values. These results were then used as input for the LCA to determine the environmental impact of the three technology scenarios and compare them with business-as-usual alternatives. For a better overall understanding of the process chain, all results were scaled to the functional unit of 1 metric ton char.

4.1. Process Simulation in Aspen Plus

4.1.1. Simulation Settings

Following the approach described in literature (Elkhalifa et al., 2019), the process was simulated continuously in a steady-state and under constant pressure (isobaric). The electrolyte NRTL (non-random two-liquid) model with Redlich-Kwong equations were used to calculate the aqueous and mixed solvent process. Furthermore, heat losses were neglected, and a uniform temperature distribution was assumed. To simulate the three technology scenarios, different reactor types provided by Aspen Plus were modelled,, the links between them are shown graphically in the process flowsheet (Figure 7) and described in the following text. The process was designed to produce a quantity of 6,4 million metric tons in the 8.000 annual operating hours. The proximate and ultimate analysis of the feedstock as well as the used reactor types and their specific inputs can be found in Appendix 7.1.

In the baseline scenario the digested sludge from anaerobic digestion (FS-Sl) is first directed into a mechanical filter (SP1-F1) to squeeze the free water out of the sludge. In the Aspen model, the wastewater (WS-WW) from this process is not considered further. The moist filter cake (S-MFC) is dried in a dryer (SP1-DR1), whereby the remaining water evaporates. The dried filter cake (S-DFC) is decomposed in the slow pyrolysis reactor (SP1-R1) into char, bio-oils and gases. Larger char particles can be taken directly from the reactor (PS-Char), but smaller char particles are carried away by the gas flow of the volatiles (S-GOP) and have to be separated from it in a cyclone (SP1-SP1). In the basic model, both the bio-oils and the gases (S-GO) are burnt in the combustion chamber (SP1-FU1) to generate the heat required for the processes. The oxygen required for the combustion process is provided by fresh air (FS-Air), which is compressed to a low overpressure by a compressor (SP1-C1). The amount of oxygen has been adjusted so that a complete combustion takes place and that there is no carbon monoxide in the exhaust gas stream. The hot exhaust air is directed through 2 heat exchangers (SP-E1 and SP-E2) to generate first very high-pressure steam (HS-VHP) with a temperature of 500°C and then low-pressure stream (HS-LP), a lower grade steam at 150°C. While high-temperature steam is required for the pyrolysis process, the drying process can be carried out with a lower steam temperature, allowing better use of waste heat energy. The water (S-H2O) required for steam generation is pressurized to 3 bar by a water pump (SP1-P1). The purification of the exhaust air (WS-EM) is not modelled in Aspen, as the calculated emissions do not correspond to reality due to the idealized calculation of the combustion reactions.

In the Torwash scenario the digested sludge is filtered to a thick sludge (S-TS). This thick sludge is than treated in the Torwash reactor (SP1-E3) and afterwards filtered again (SP1-F2) before a filter cake with a higher solid content is obtained (green section in Figure 7). For simplification reasons, the Torwash process was modeled as a heat exchanger that first heats up the feedstock and then cools it down again. The treatment in the Torwash reactor enables the mechanical filter to further reduce the moisture content of the filter cake, so that less energy is required for the drying.

In the bio-oil scenario modelled in Aspen, the volatile gas stream is directed through a condenser column (SP1-T1) (blue section in Figure 7). This column is based on the work of Ibarra-Gonzalez, has 15 stages and operates with a quench water temperature of 60°C (Ibarra-Gonzalez & Rong, 2019). Thereby the temperature is cooled down to such an extent that the condensable bio-oils (PS-Oil) are separated from the non-condensable gases (S-G). The needed quench water (S-H2O) is preheated by the liquid phase leaving the condenser (PS-Oil) in another heat exchanger (SP1-E4). As the separation of the bio-oils from the quench water can be achieved via a retention tank without further energy input, the separation is not modeled in Aspen. The non-condensable gases (S-G) are directed into the combustion chamber and burned there.

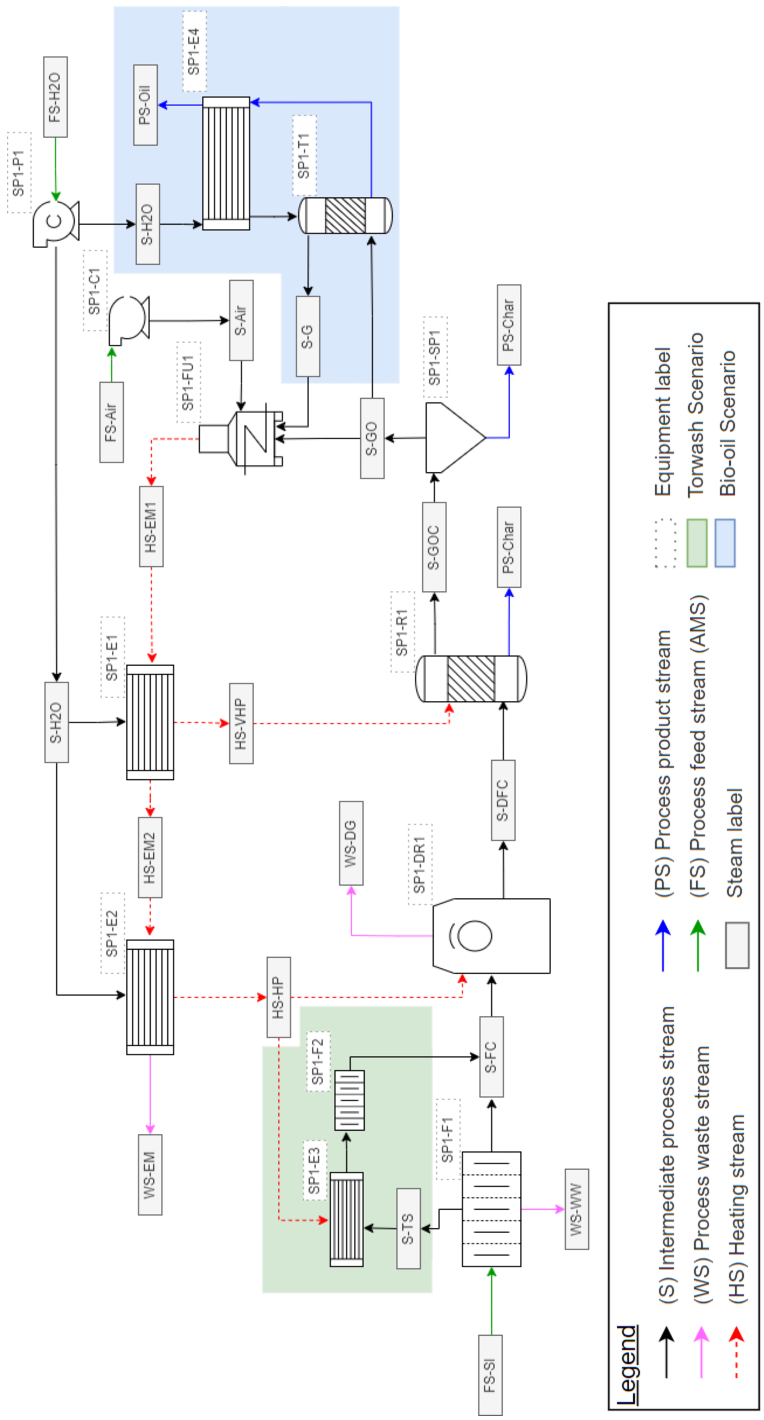

Figure 7 Flow chart of the process simulation modeled in Aspen Plus

4.1.2. Energy consumption of the Mechanical Dewatering

The filtration of solid particles from liquids can be simulated in Aspen Plus with two different technologies: filter press and centrifuge. However, the energy consumption of both simulations is significantly lower than described in the literature (Figure 8). This can be explained by the simplification made in the simulation that no interactions take place between the particles and the water. Therefore, the water can be pressed out of the sludge with less energy. In practice, the interactions mean that a higher solid content causes higher energy consumption. (Jules van Lier, 2023b; Sludge Processing, 2020).

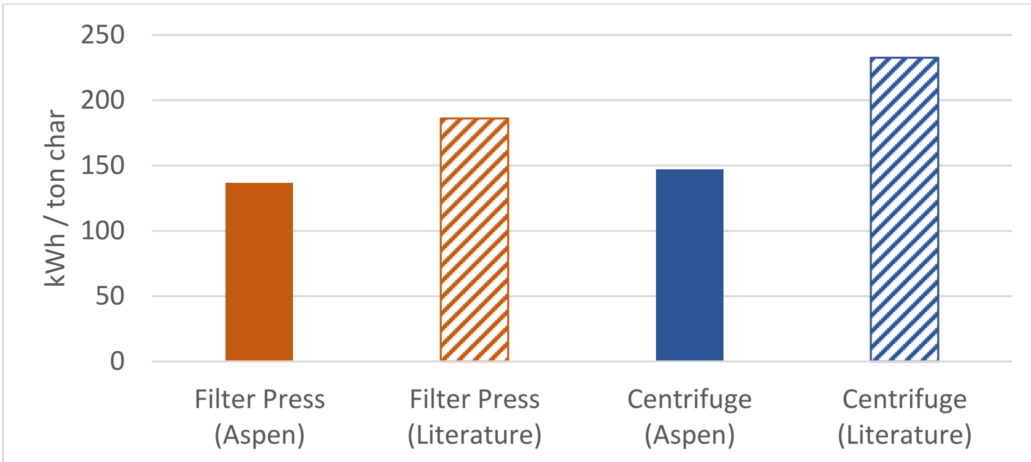

Figure 8 Energy consumption of different dewatering technologies compared with their respective literature value.

4.1.3. Energy Consumption of the Dryer

Although industrial dryers often only achieve a solid content of 90% (Huber SE, 2023), the dryer in Aspen was simulated so that all the remaining water evaporates. This simplification has no impact on the overall energy balance, as the remaining 10% would otherwise have to be evaporated in the slow pyrolysis reactor. The same simplification that led to lower energy consumption for the filter also leads to lower energy consumption for the dryer. While the Aspen simulation assumes a thermodynamically ideal drying process with 100% efficiency, industrial dryers reach only an efficiency of 75 to 90% (Cheng et al., 2020; Huber SE, 2023). Figure 9 shows not only the effect of different dryer efficiencies but also the influence of the solid content (SC) in the filter cake on the energy demand of the dryer. The red dashed line shows the potential energy savings of the torwash scenario, where the solid content is increased from 35% to 50%.

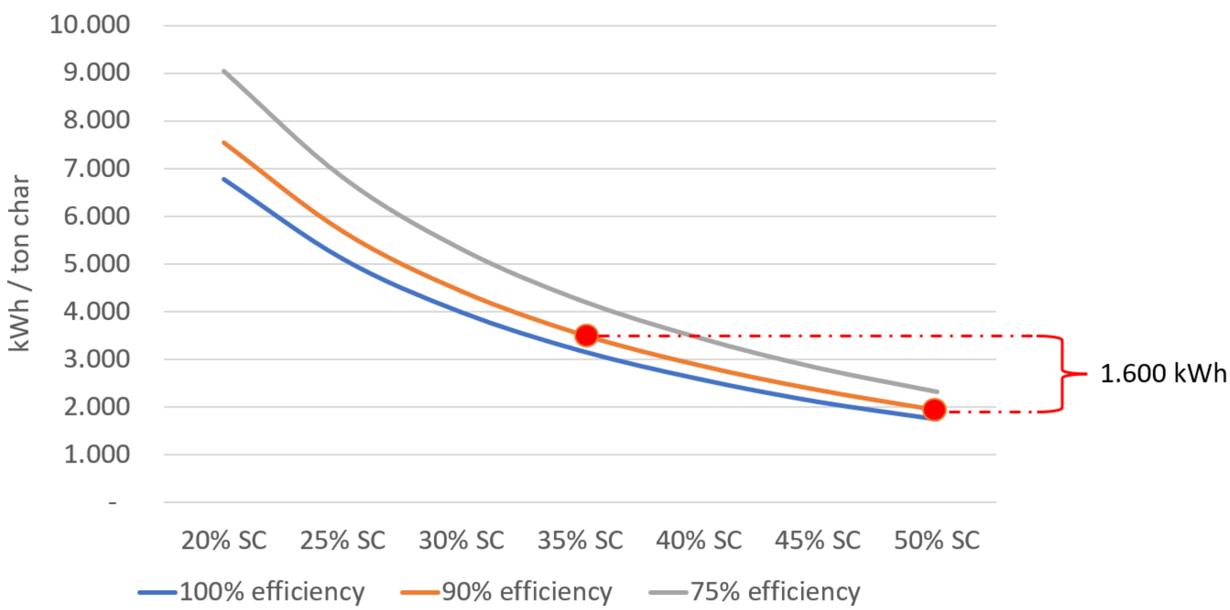

Figure 9 Energy consumption of the drying process dependent on the efficiency and solid content (SC) of the filter cake

4.1.4. Energy consumption of the Torwash Process

In the Torwash process, thick sludge with 20% SC is thermally treated, before a second filtration step can increase the SC to 50%. To heat up the thick sludge to temperatures of 200°C over 10 MW thermal energy is required. However, in an ideal thermodynamic scenario heat exchangers could heat up the inflow with the thermal energy of the outflow, reducing the theoretical steady state energy consumption to 0. In practice, however, heat losses occur due to insufficient insulation or limited heat transfer capacity in the heat exchanger. In Figure 10, these heat losses are plotted as a percentage of the initial energy consumption. The energy saved due to the higher solid content in the drying step amounts to 1.600 kWh per ton char. This means that the heat losses of the Torwash process must not exceed 16% of the initial energy requirement in order for the process to result in energy savings.

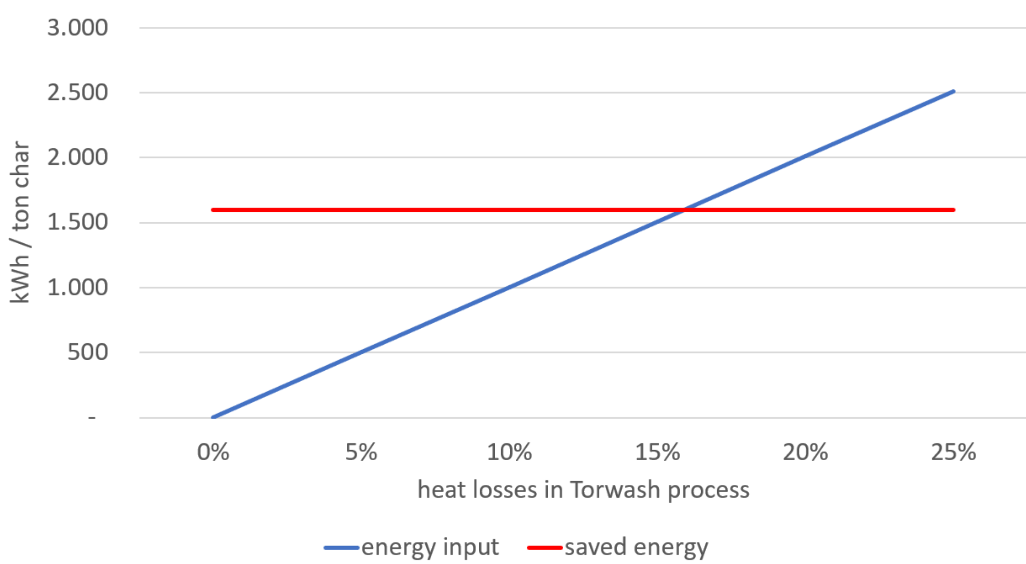

Figure 10 Energy consumption of the Torwash process respective to its heat losses.

4.1.5. Energy consumption of the Slow Pyrolysis Reactor

The energy consumption of the slow pyrolysis reactor is made up of two components, the energy required to heat the dried filter cake to the reactor conditions (from 100 to 400°C) and the energy needed for the endothermic reaction. The reaction energy can be further split into the energy required for the formation of the gases, bio-oils and char. The results of the Aspen simulation (Figure 11) show that the ash content of the feedstock has a significant influence on the reaction energy. This can be explained by the composition of the ash, which consists mainly of metals which are inert at the reaction temperatures of 400°C. Since the ash remains completely in the char, it can be assumed that the reaction energy for the formation of the gases and bio-oils remains constant, and the ash content only influences the reaction energy needed for the formation of the char. The sewage sludge investigated in this study has with 86% a high ash content, resulting in a relatively low energy requirement of 639 kWh per ton of char for the slow pyrolysis reactor.

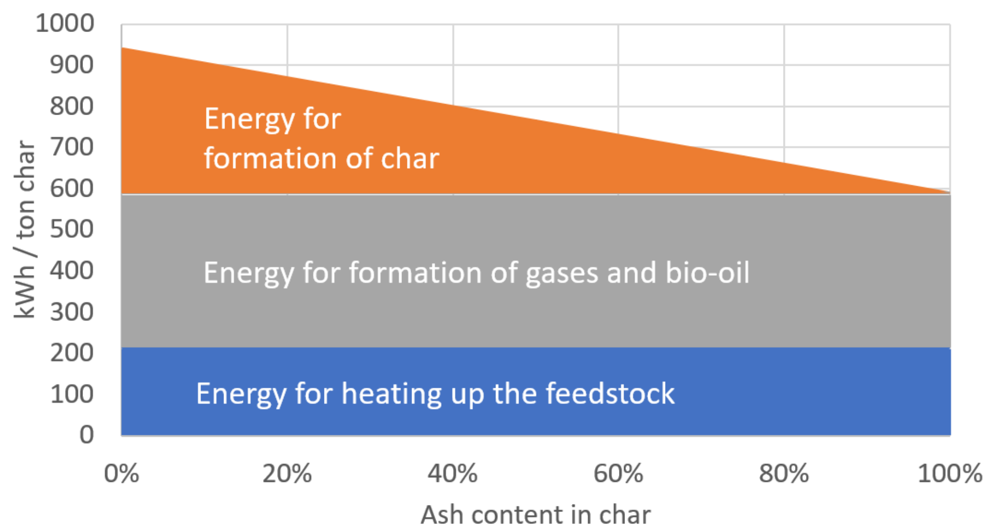

Figure 11 Energy consumption of the slow pyrolysis reactor allocated to the energy consumption of the heating of the feedstock and the energy consumption for the formation of the gases & bio-oils and the char

While larger char particles can be removed directly from the reactor, smaller particles are carried along by the gas stream. These can be separated from the gas stream in a cyclone without the need for further energy.

4.1.6. Energy Consumption of the Condenser

The in Aspen simulated condenser achieved a separation is displayed in Figure 12. For the fraction of non-condensable gases (Methane, carbon dioxide, carbon monoxide, and hydrogen) the condenser achieves a high separation rate of 99,7%. However, only 90% of the bio-oils can be condensed. The fraction of non-condensable bio-oils consists mainly of three components: 2-ethylhexanol, pyridine and styrene (Figure 12). Investigations revealed that their respective boiling temperatures of 180 °C, 115 °C and 145 °C are significantly higher than the temperature of the condenser (PubChem, 2023), and thus they theoretical should condense. This leads to the assumption that there is an error of unknown origin in the simulation. In order to prevent the error from propagating, a perfect separation of the condensable bio-oils and non-condensable gases is assumed for the following processes. Nevertheless, the simulation showed that the energy required for heating the quench water can be extracted from the discharge of the liquid phase in a heat exchanger. Therefore, the energy consumption of the condenser is limited to the pumping power for the quench water.

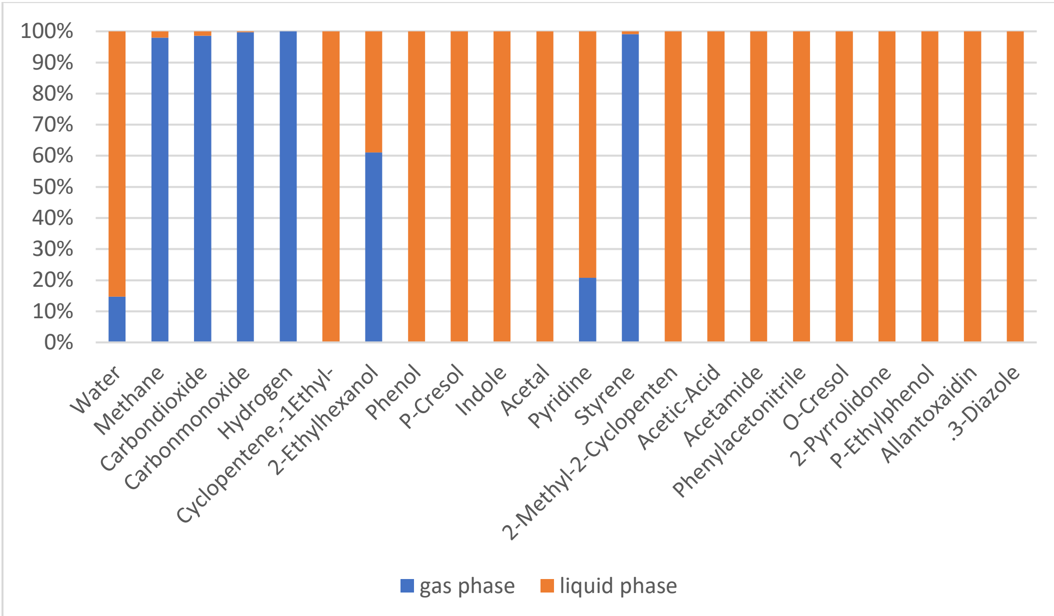

Figure 12 Composition of the gas and liquid phase

4.1.7. Thermal Energy Balance

In the baseline scenario, the energy generated by the combustion of gases and bio-oils exceeds with 8.800 kWh the demand for heat energy with 4.180 kWh per ton char. Since heat energy is also needed in other H2Steel processes associated with the following chemical leaching, which are not considered in this study, the excess heat is reserved for these. For a comparison of the different technology scenarios, the Torwash and Bio-Oil scenarios need to provide this same amount of excess heat energy. In this study, this heat is generated using an electric heater.

The thermal energy balance in Figure 13 shows that drying is the largest energy consumer for the baseline and bio-oil scenario with 3.540 kWh per ton char. In the Torwash scenario this demand is reduced to 1.670 kWh per ton char due to the higher solid content of the filter cake. Instead, the Torwash process is a significant energy consumer with 4.650 kWh per ton char. In the bio-oil scenario the same amount of heat is required as in the baseline scenario, but only 42% of it is produced via the combustion of the pyrolysis gases, the rest is supplied via electricity.

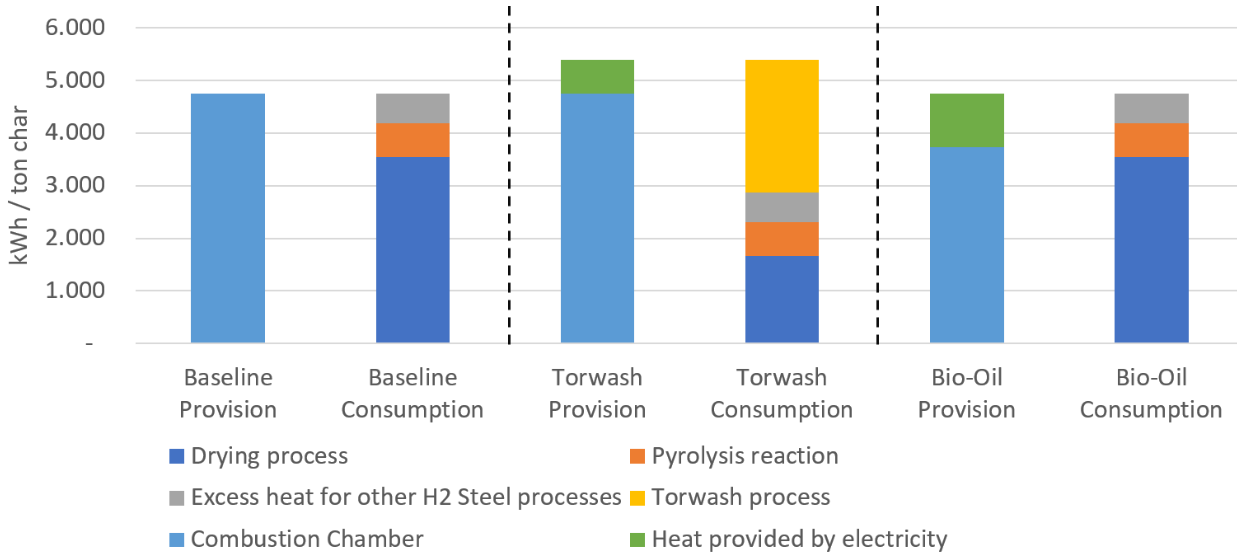

Figure 13 Thermal energy balance of the process simulation

4.1.8. Electric energy consumption

In the baseline scenario, the filter press consumes 186 kWh per ton char, making it the largest energy consumer. In the Torwash and bio-oil scenario, the electrical consumption for the heat production is significantly higher with 640 and 1.015 kWh per ton char, respectively. However, with the use of high efficiency heat pumps, the electricity demand for the process heat can be reduced by a factor of 4 (Figure 14).

In the bio-oil scenario, the energy consumption of the air compressors is 55 % lower. This is because the bio-oils are not burned and therefore less air is needed for combustion. The electricity savings for the water pumps are partly used up by the additional process water needed for the condenser, so the reduction is only 25 %.

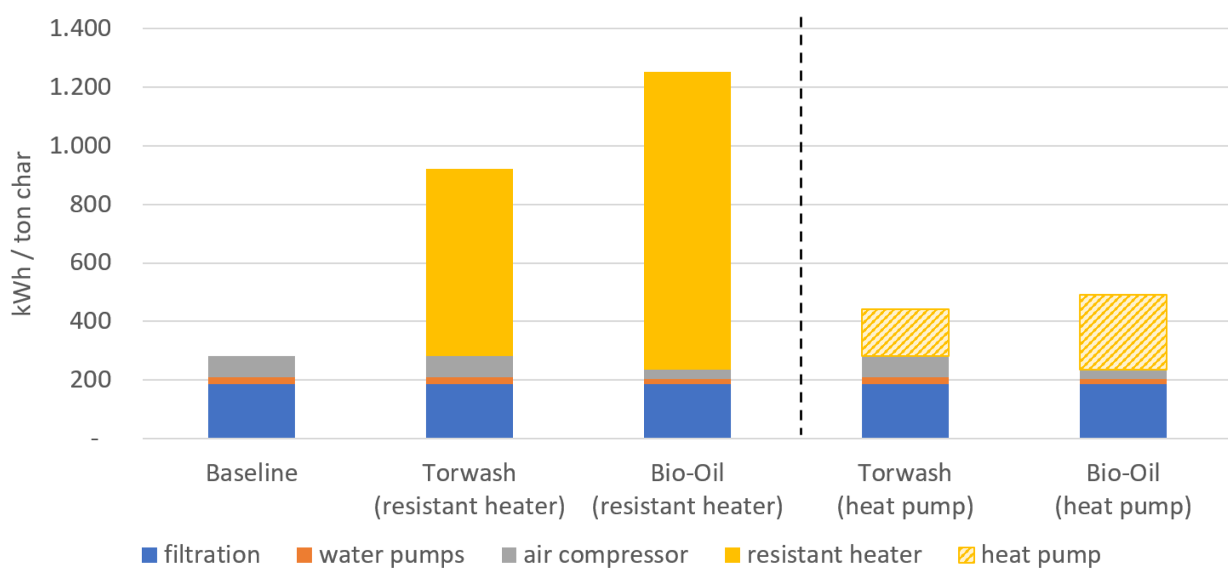

Figure 14 Electric energy consumption

4.2. Results of the Environmental Impact Assessment (LCA)

4.2.1. Life Cycle Inventory Analysis

The product system investigated in the LCA is based on the process modelled in Aspen. In addition, the LCA also takes into account background processes such as the treatment of the filter water, the production of flocculants for the filter process and chemicals for flue gas cleaning. The processes considered, as well as the system boundaries, are illustrated graphically in the flow chart (Figure 15). The specific inputs for the technology scenarios can be found in the inventory table in Appendix 7.2.

The feedstock enters the system without any environmental burden, as the digested sludge is a waste product of anaerobic digestion. As the slow pyrolysis process also takes place on the site of the wastewater treatment plant, no transport is required. For the energy consumption of the filtration, the literature value of the filter press was used since both the energy requirement and the quantity of flocculants are lower than for the centrifuge. As there is no EcoInvent process of the production of polyacrylic ether, the most used flocculant, the production of acrylic acid, a base material for polyacrylic ether, was used as a substitute. The filtration water is processed in a further treatment step to reduce the high phosphorus and nitrogen content of the water. Since the wastewater has already undergone biological purification, only the last section of the wastewater purification process needs to be carried out. For this, the EcoInvent process "treatment of wastewater from anaerobic digestion of whey" was used as an approximation for the energy consumption and environmental emissions. For the drying stage, a solid content of 35% achievable by filter presses and a thermal efficiency of 90% corresponding to the latest generation of industrial dryers were assumed. The exhaust gas purification is based on the EcoInvent process “Incineration of Sewage Sludge”. However, the inflows are adjusted, so that only relevant chemicals for purification are considered. Furthermore, heavy metals and ash were removed from the emissions, as they remain in the char.

As described in chapter 2.11, the Torwash scenario builds on the baseline scenario and differs only in the preprocessing. As no precise data on the heat losses of the Torwash process are available, a conservative approach was chosen and a heat loss of 25 % was assumed. The process heat required for this comes partly from the savings in the drying process and partly from an electric heater. For the conversion of electrical energy into thermal energy, two technologies are examined in the LCA, resistant heaters and industrial heat pumps. While resistant heaters convert 100% of the electrical energy into heat energy, heat pumps used for this LCA have an efficiency factor of 400%. In addition, it is assumed that the two filtration steps, which each require less energy, together require the same amount of energy as the filtration step of the baseline scenario.

The bio-oil scenario differs from the baseline scenario only in the utilization of the condensable bio-oils. Since the fault in the separation between the condensable and non-condensable gas fraction could not be found, an ideal division of the two phases is assumed. To compensate for the heat that would otherwise be generated during the combustion of the bio-oils, an electric heater is used. The multifunctionality created by the co-product bio-oil is solved with a system extension.

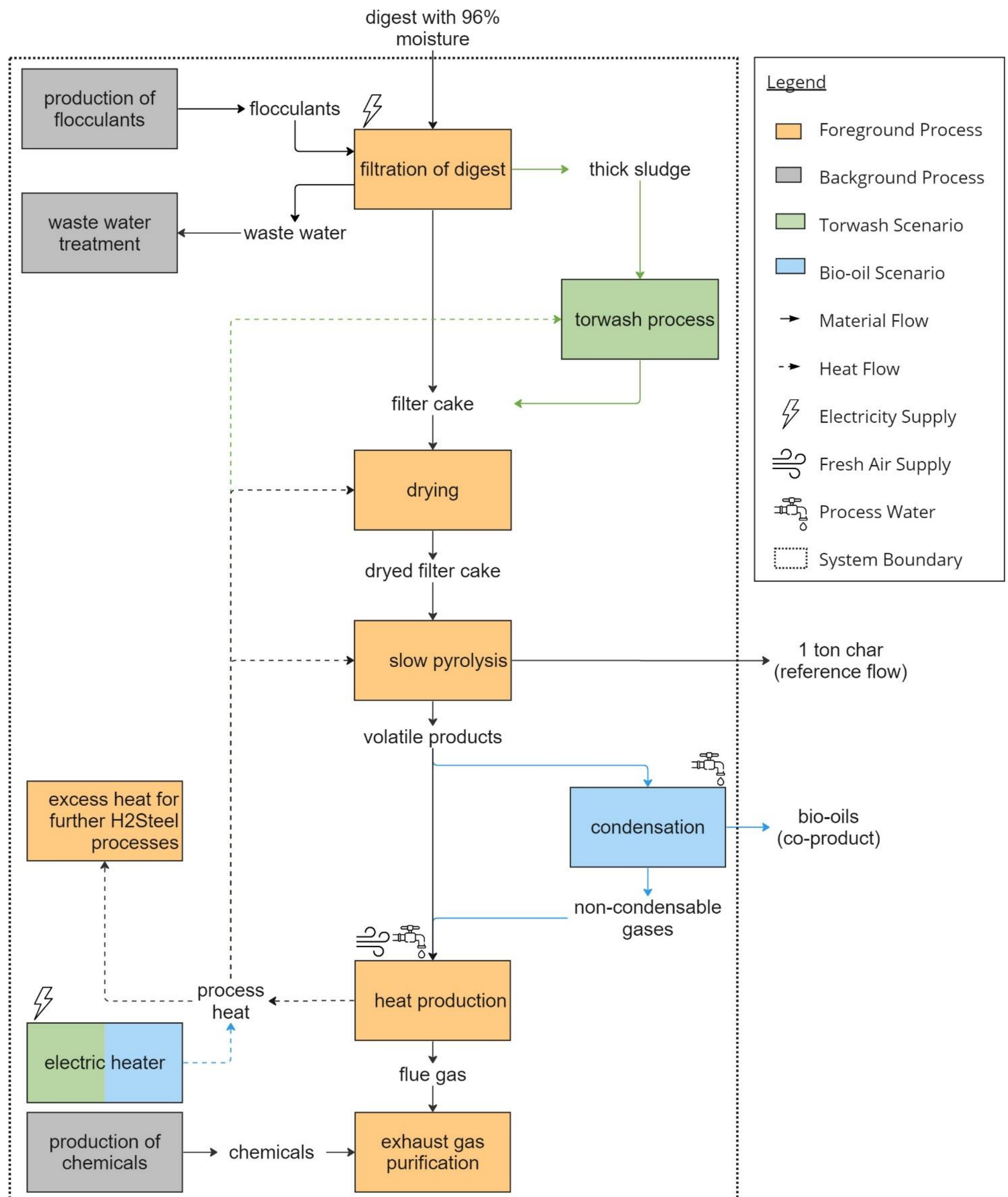

Figure 15 LCA flow chart of the base line slow pyrolysis process (orange and grey) as well as the two alternatives the Torwash scenario (green) and the bio-oil scenario (blue)

The business-as-usual scenario must fulfil the two functions of the slow pyrolysis process: (I) end-of-life treatment of sewage sludge and (II) production of a coal alternative. As an end-of-life alternative, the EcoInvent background process "treatment of sewage sludge in municipal incineration with heat recovery" was chosen. This process requires a filter cake with 73% moisture as input. Since the feedstock for all investigated alternatives is the same, a preprocessing filtration step of the sludge with the necessary background processes is also considered. The EcoInvent market for hard coal in Europe is used as an alternative to char production. The market includes mining and transport required from various coal exporting countries. The amount of coal was chosen so that the heating value corresponds to that of a ton char. As the excess heat that is considered in the other alternatives ultimately remains in the system, the business-as-usual scenario does not have to provide this energy.

In accordance with the system expansion, the business-as-usual alternative for biofuel also has to take into account the provision of an oil alternative (Figure 16). For this, petroleum was used, which, like the bio-oils, requires a further refining process. The amount of petroleum was chosen so that the heating value corresponds to that of the bio-oil.

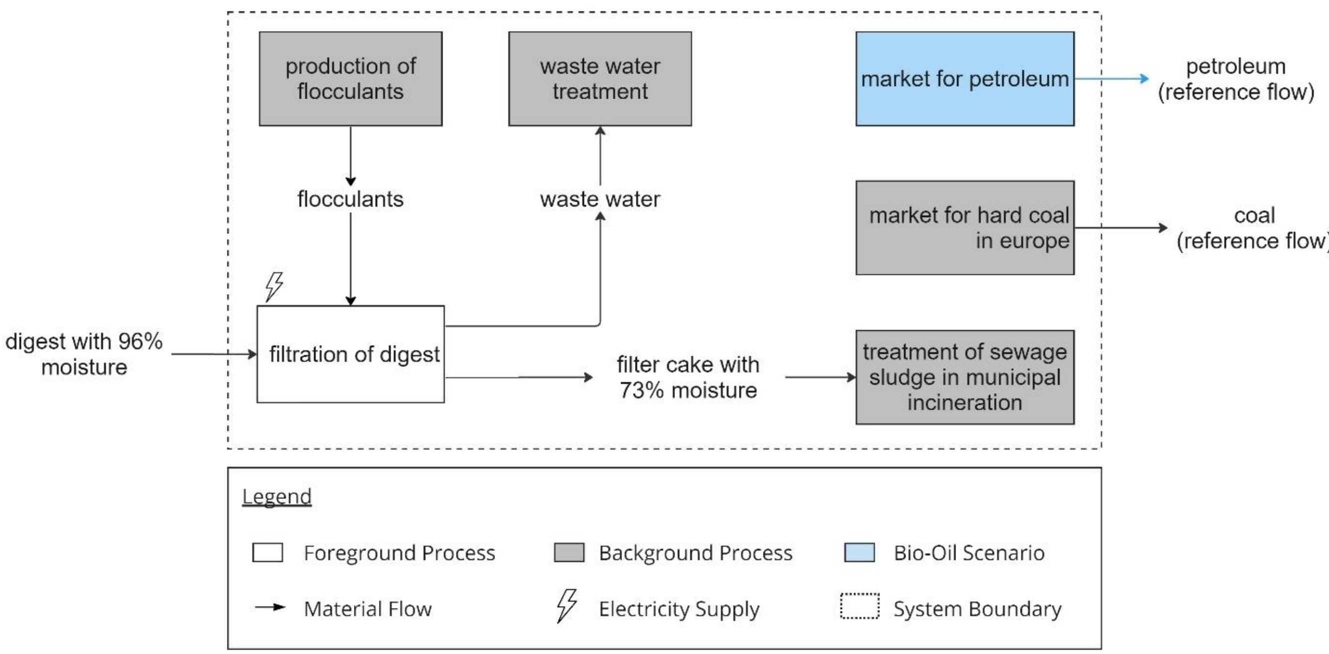

Figure 16 LCA flow chart of the business-as-usual scenario and the business-as-usual alternative for the bio-oil scenario (blue)

4.2.2. Life Cycle Impact Assessment (LCIA)

The LCIA processes the results of the inventory analysis into contributions to relevant impact categories. For this, a classification and characterization of the inventory table is necessary. In the classification, the individual interventions (emissions, resource extractions and land use) are subdivided into the different impact categories. To account for the different impacts of the interventions, these are multiplied with specific characterization factors before they aggregated into a single score (Guinée, 2002).

In accordance with the European framework of the H2Steel project, this report uses the impact family selected by the EU. The environmental footprint impact family developed in 2016 is subject to the EN15804 standard and consists of 19 impact categories (European Commission, 2016). Of these, the 5 relevant impact categories (I) climate change, (II) acidification, (III) freshwater ecotoxicity, (IV) eutrophication of freshwater and (V) eutrophication of marine water were selected, to assess the environmental performance of the different alternatives. While the baseline and the Torwash scenario have to be compared with the business-as-usual alternative, the bio-oil scenario has to be compared with the business-as-usual bio-oil alternative due to the system expansion. Figure 17 shows the respective results normalized to the largest value in case a resistant heater is used.

The impact category **climate change** uses the characterization method global warming potential (GWP100). This method was developed by the Intergovernmental Panel on Climate Change (IPCC) and defines the global warming potential of various greenhouse gases over a 100-year time horizon (IPPC, 2007). A contribution analysis of the impacts of the different scenarios will be presented in more detail in the next chapter.

To calculate the **acidification** impact, the accumulated exceedance (ae) model developed by (Seppälä et al., 2006) is used. This assesses how many hydrogen ions are formed from the mineralization of NO_x, NH_3 and SO_x emissions. The resulting protons contribute to the acidification of soil and water which can result in forest decline and lake acidification. The results for acidification in Figure 17 show significant higher values for the Torwash and Bio-oil scenario. This can be explained by extra emissions caused by the electricity production.

For **ecotoxicity of freshwater**, the USEtox characterization method developed by Rosenbaum et al., (2008) is used. The method comprises characterization factors for several thousand substances that can cause harm to individual species or a change in the structure and function of an ecosystem. The impact of all scenarios does not show any significant deviations (Figure 17). This is because for all scenarios at least 98% of the emissions for this impact category come from the treatment of the filtration wastewater and these have only a slight variation over the different scenarios.

The impact categories **freshwater** and **marine eutrophication** are based on the EUTREND model developed by Struijs J. (2009). The EUTREND model is based on the assumption that phosphorus emissions are decisive for eutrophication of freshwater, while nitrogen emissions are the determining factor for eutrophication of the sea. This is justified by phosphorus often being the limiting nutrient in inland waters, while in the marine environment the so-called Redfield ratio is more important. This ratio refers to the composition of aquatic phytoplankton ($C_{106}H_{263}O_{110}N_{16}P$), which requires 16 times the amount of nitrogen per phosphorus (Redfield et al., 1963). Therefore, nitrogen is considered a bottle neck for marine eutrophication. In addition, Struijs emphasizes the role of municipal wastewater treatment plants as the main source of phosphorous and nitrogen nutrients entering aquatic systems. While the impacts for marine eutrophication are relatively constant, the business-as-usual scenarios show significantly higher values for freshwater eutrophication (Figure 17). This is because some of the phosphorus dissolved in the sludge remains in the char, while in the business-as-usual alternative it is burned and thus released into the environment.

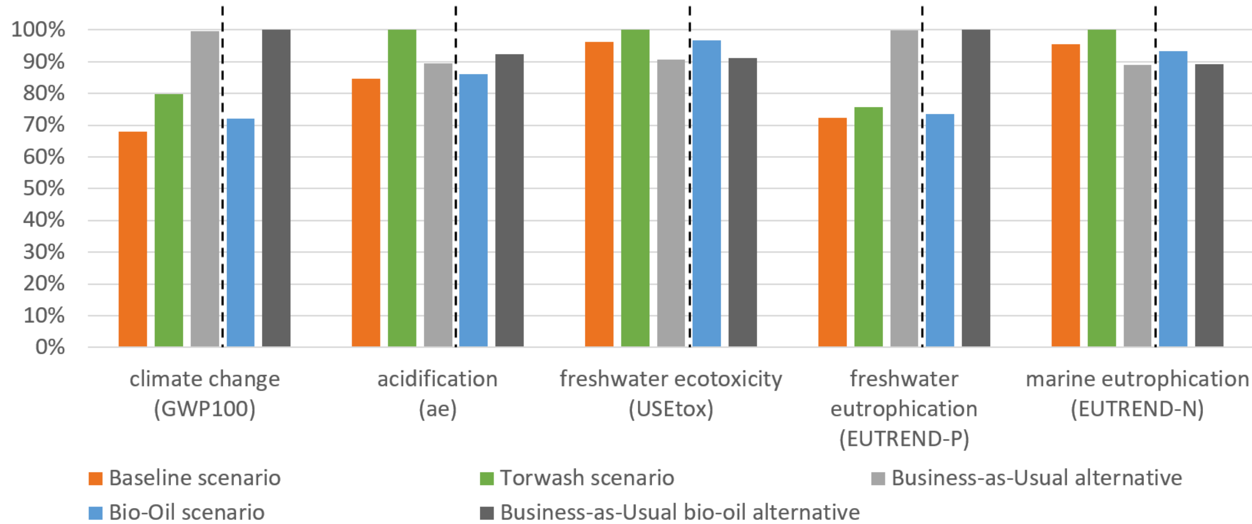

Figure 17 Environmental footprint (EF v3.0) iImpact assessment of char production via different technology scenarios compared to business-as-usual alternatives (normalized to the highest value)

4.2.3. Interpretation of the LCIA results

For a more detailed analysis of the results, a process and environmental flow contribution is carried out. Additionally, the most important assumptions are examined in the sensitivity analysis in order to determine the robustness of the results. A comparison to literature values, which would further strengthen the validity of the results, was not possible. This is because the LCAs described in the literature either have a different scope and thus a different process chain (Cheng et al., 2020; Peters et al., 2015a), or report incompletely on the assumptions and inventory table (Barry et al., 2019).

4.2.3.1. Contribution Analysis

Figure 18 shows the detailed process contributions for the impact category climate change. The comparison between the baseline scenario and the Torwash scenario clearly shows that the 25% heat losses in the Torwash process exceed the energy savings in the drying stage. This leads to CO_2eq emissions of the Torwash scenario being even higher than those of the business-as-usual alternative. Both business-as-usual scenarios are determined by the end-of-life treatment of the sludge. More precisely, 24% of the total emissions are due to the filtration process and the purification of the filter water, 74% to the combustion of the filter cake, and only 2% to the mining and transport of the coal. The additional supply of oil in the conventional bio-oil alternative adds only 0,4% to this. The CO_2eq emissions of the bio-oil scenario lies between those of the baseline and the Torwash process. The largest contributor is with 452 kg CO_2eq the production of excess heat with electricity.

The detailed process contributions for the other impact categories can be found in appendix 7.3.

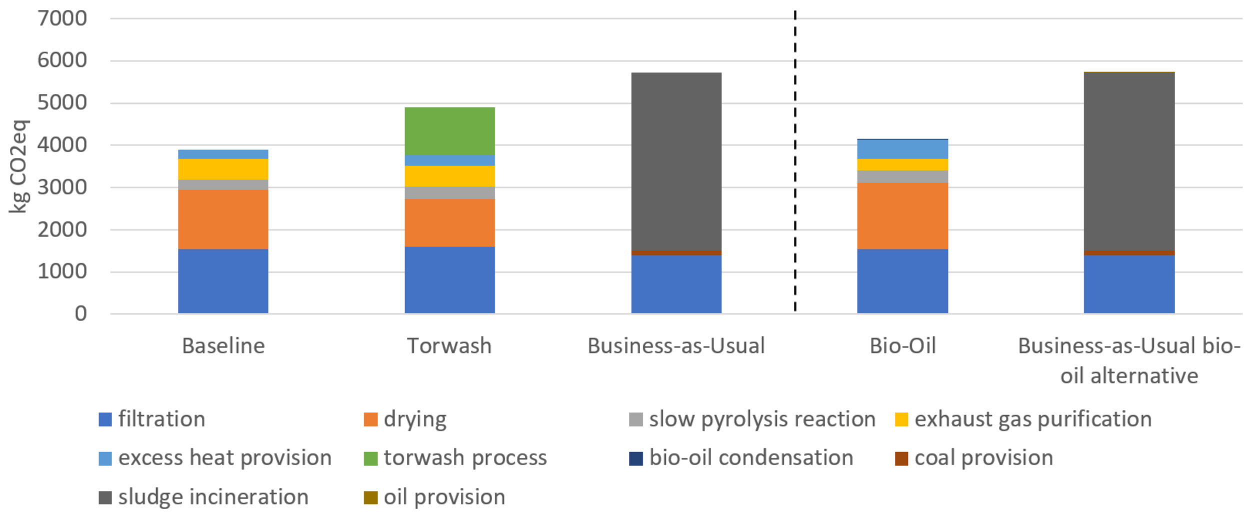

Figure 18 Process contributions to the impact category climate change (GWP100) of the different scenarios

The environmental flow analysis in Figure 19 provides further information on the origin of the emissions. While the baseline scenario emits 2.580 kg of non-fossil CO_2, i.e. CO_2 that was previously absorbed by plants, the business as usual alternative emits 3.510 kg. This difference can be explained by the carbon stored in the char. In the bio-oil scenario, the difference is even larger because additional carbon is stored in the bio-oils. In contrast, the higher electricity consumption of the bio-oil scenario leads to significantly higher fossil CO_2eq emissions. These originate from electricity production, which in Italy is still 68% based on fossil fuels (EcoInvent, 2021). In the absence of process-specific data, flue gas cleaning and associated emissions were copied from the business-as-usual scenario. Therefore, nitrous oxide emissions from this process are constant for all scenarios except the bio-oil scenario. In this scenario, flue gas cleaning was reduced in proportion to the volumetric exhaust gas flow rate. This adaption results in 38% lower dinitrogen oxide emissions.

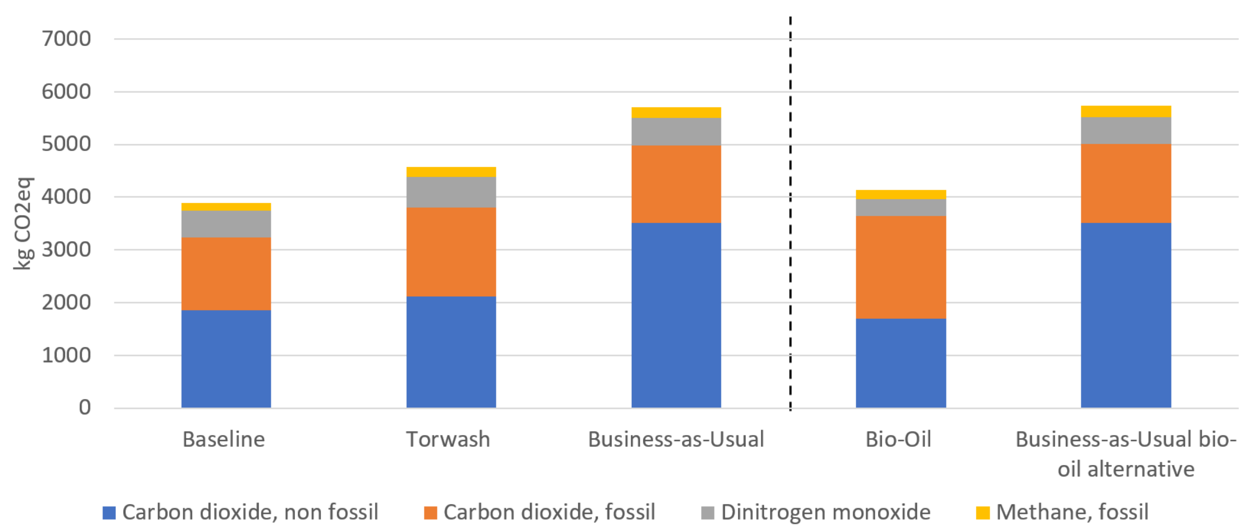

Figure 19 Environmental flow contributions to the impact category climate change (GWP100) of the different scenarios

4.2.3.2. Sensitivity analysis

As the contribution analysis shows the production of process heat which for the bio-oil scenario is partly based on electricity is one of the largest emitters. At the same time, the electricity mix and thus its emissions vary greatly in the European countries. For this reason, the sensitivity analysis examines how the country-specific electricity mix affects the CO_2eq emissions of the baseline and bio-oil scenarios. As the Torwash scenario led to higher environmental impacts in all impact categories than the baseline scenario, it is considered in the sensitivity analysis. For an easier interpretation of the data, the map extension of Excel is used to display the difference between the baseline and the bio-oil scenario according to Equation 4 on a European country map.

$$Difference = Climate\ Change\ Impact\ Baseline - Climate\ Change\ Impact\ BioOil \qquad [4]$$

If a country has a positive value (blue), the bio-oil scenario is to be preferred. On the other hand, a negative value (orange) indicates a preference for the baseline scenario. The results in Figure 20 show that in countries like Finland or France, whose electricity mix have a relatively low CO_2 intensity, the production of bio-oils is to be preferred. In countries that still have a relatively high CO_2 intensity in the electricity grid, such as Poland or the Czechs, it is less environmentally damaging to burn bio-oils for heat production than to use electricity for this purpose.

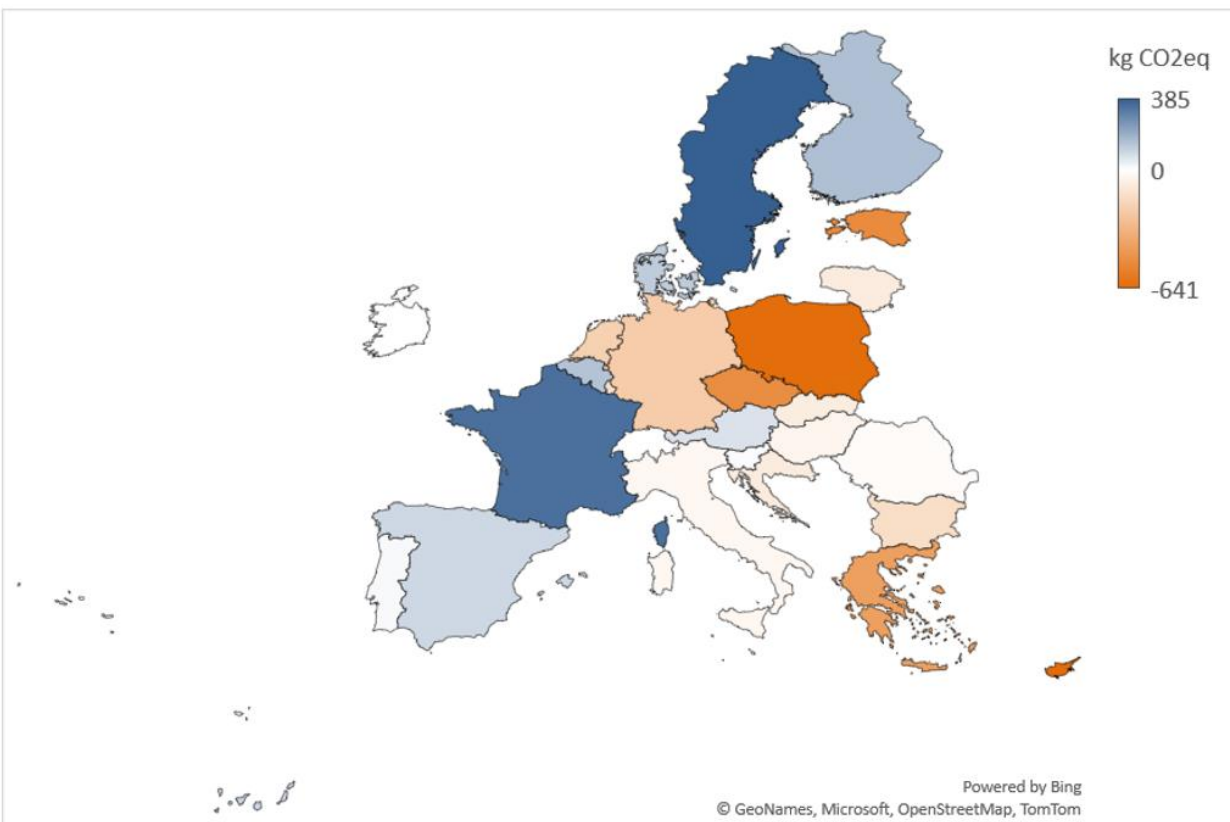

Figure 20 Visualization of the difference between the baseline scenario (orange) and the bio-oil scenario with resistant heater (blue) for the impact category climate change (GWP100). The coloring indicates which scenario has a lower environmental impact in a particular country.

If for the provision of process heat a more efficient heat pump is used, the emissions of the bio-oil scenario are reduced significantly. The reduction is so large, that in all EU member states, the bio-oil scenario would emit less CO_2eq emissions than the baseline scenario (Figure 21).

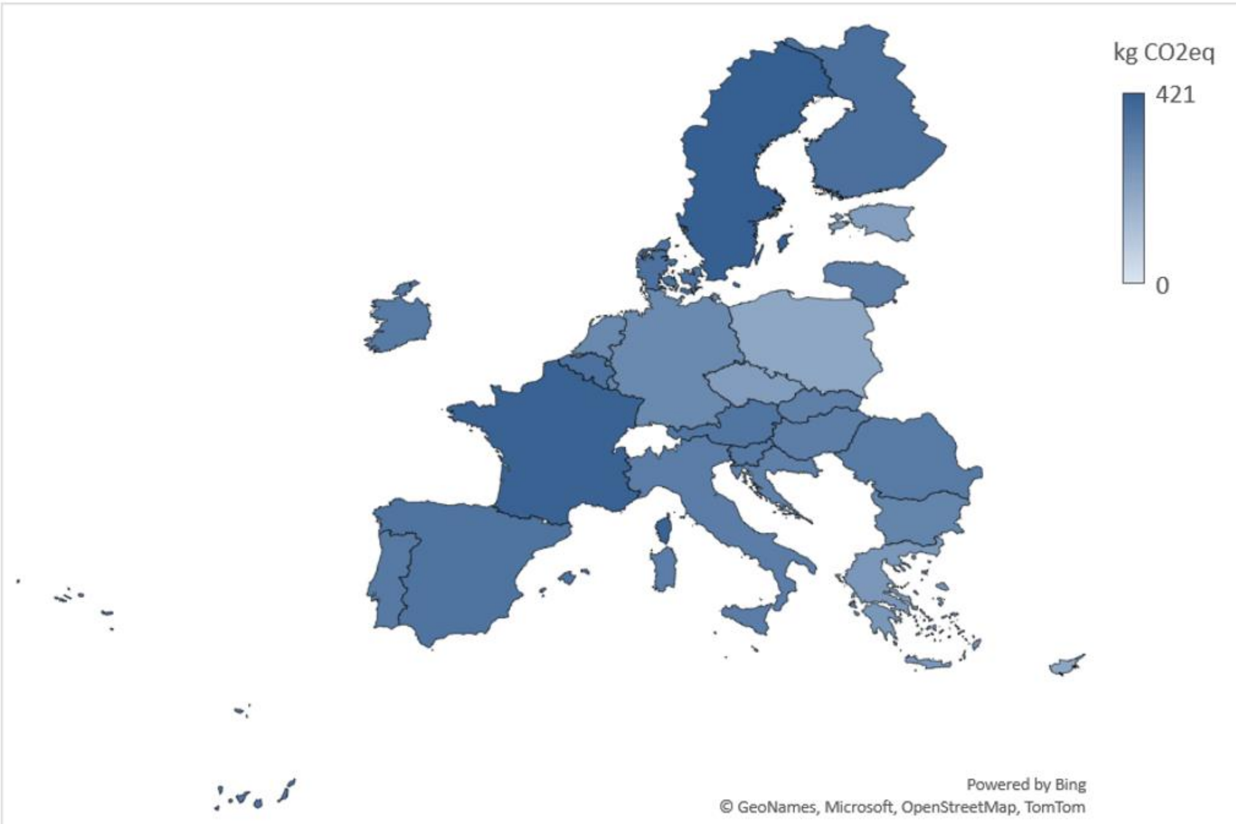

Figure 21 Visualization of the difference between the baseline scenario and the bio-oil scenario with heat pump (blue) for the impact category climate change (GWP100).

The use of a heat pump results also in a significant reduction in the other impact categories. Figure 22 shows these changes for the Italian electricity mix. The bio-oil scenario now has the lowest environmental impact in almost every impact category. Only in the impact category freshwater ecotoxicity the bio-oil scenario with a heat pump has a 2% higher impact than the baseline scenario.

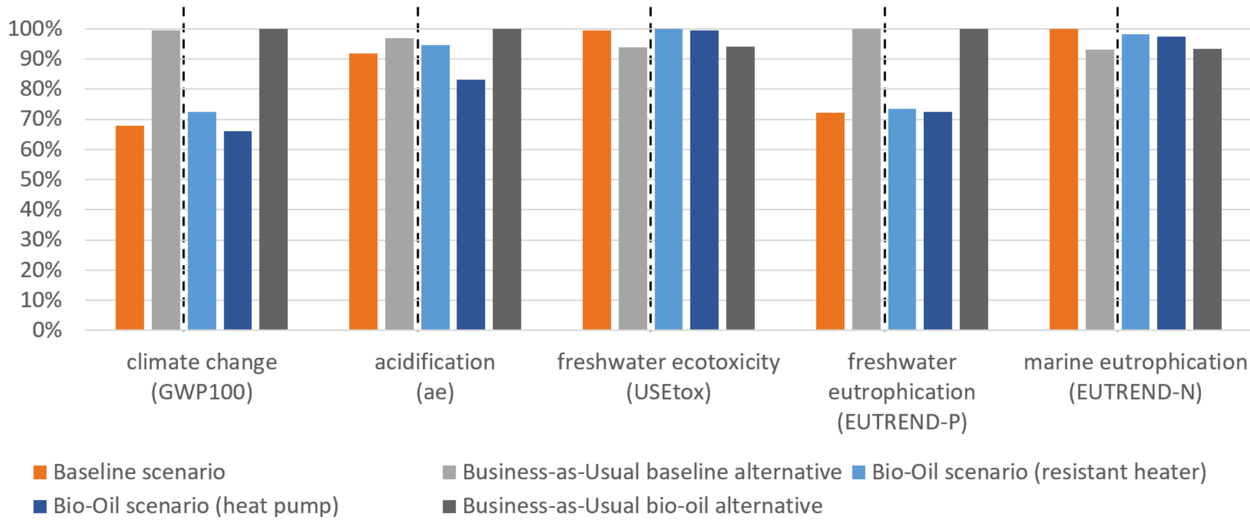

Figure 22 Environmental footprint (EF v3.0) impact assessment of char production via different technology scenarios and different electric heaters (normalized to the highest value)

4.2.4. Results of the ex-ante LCA

The emissions are even further reduced when future scenarios are considered. In Figure 23, the baseline and bio-oil scenario with heat pump are compared on three different time horizons: 2021, 2030 and 2040. For this, the more conservative IAM model was used, which leads to 2.5°C warming by 2050. As the IAM model does not differentiate the different European electricity mixes, the 2021 scenario is here also modeled with a European electricity mix.

It is evident that the two impact categories climate change and acidification have a strong reduction from 2021 to 2030, followed by a moderate reduction towards 2040. The strong reduction by 2030 is mainly due to the shutdown of CO_2-intensive coal-fired power plants. Figure 23 also shows that the difference in the impact categories climate change and acidification will continue to grow between the baseline and the bio-oil scenario in the future. The other 3 impact categories show only a slight improvement, maximal 6% in the impact category freshwater eutrophication. This is due to the fact that the future scenarios do not yet include technological improvements for the main polluter, the treatment of filtration water.

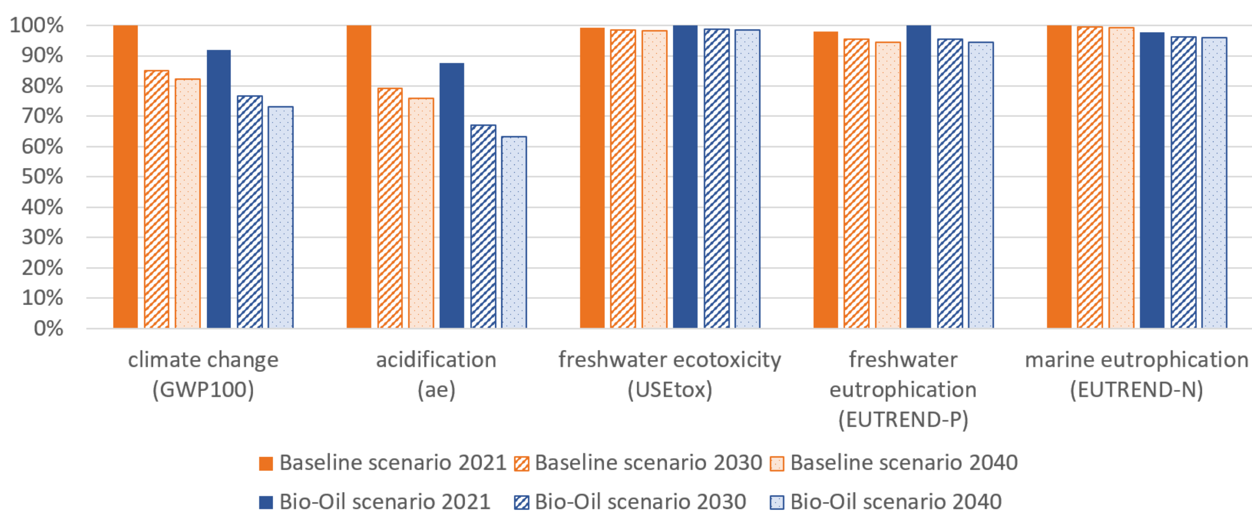

Figure 23 Environmental footprint (EF v3.0) impact assessment of char production via different technology scenarios and different time horizons (normalized to the highest value)

5. Limitations and Outlook

One of the limitations of this report is that only one specific feedstock is analyzed. Yet even the sewage sludge feedstock has a wide variation in composition depending on which industries feed into the municipal wastewater system. However, this limitation could be partially mitigated by identifying the ash content of the feedstock as a significant factor in the energy consumption of the slow pyrolysis. In further work, this factor found in the simulation should be verified experimentally and coupled with the resulting heating value of the char.

Another limitation linked to the composition of the sewage sludge is the negative effect of the filtration water on the environment. Although the used sewage sludge is already biologically purified, the water contains high concentrations of phosphorus and nitrogen. Since measurement data on the exact concentration of the nutrients are difficult to find, an EcoInvent process had to be used. Additional experimental data could lead to more accurate LCA results and indicate the need for further processes to remove the nutrients from the filter water. This would not only significantly reduce the environmental impact in the impact categories freshwater ecotoxicity, freshwater eutrophication and marine eutrophication, but the extracted nutrients could also be sold as fertilizer and thus provide a further economic benefit.

Although Aspen Plus provides an estimate of steel and cement requirements for the reactor and its foundations, this analysis was not performed due to the time constraints of this study. However, it is anticipated that due to the long life of the process equipment, its contribution to the overall impact is likely to be less than one percent.

Another limitation is that the Torwash process has been simplified. While in this study only the effect on the achievable solid content of the filter cake was considered, in practice conversion processes already take place during the Torwash process that reduce the amount of organic components in the filter cake. This would not only have an effect on the emissions generated in the Torwash process itself but would also influence the production yield of the slow pyrolysis. Information on whether this has a positive effect, such as a higher yield of char due to the greater porosity of the filter cake, or whether it has a negative effect, as some of the organic content is lost, could not be found and thus represents a new knowledge gap.

A limitation of the REMIND model is its spatial resolution. It divides the world into 12 regions, and since Europe is represented as a single region, a further breakdown into individual countries is not possible for the ex-ante LCA.

This report was originally planned to be based on the experimental data from the H2Steel project. However, since this project started at the same time as this master thesis, the experiments were not yet advanced enough to integrate their results into this work. As this master thesis will be continued as part of a PhD thesis, the experimental data as well as the further processes like chemical leaching, biomethane cracking and char briquetting can be integrated into the analysis. These processes will lead to a higher overall environmental impact, but also to further economic co-products such as fertilizer and hydrogen.

6. Discussion and Conclusion

The objective of this report is to identify the most sustainable scenario to produce char from sewage sludge. For this purpose, the literature was first scoured to find the latest developments in the key processes. In the next step, these were simulated in the Aspen Plus software to obtain process-specific data on energy consumption and CO_2 emissions. In an LCA, these were combined with the necessary background processes to analyze the environmental impact of the entire value chain. Analogous to this procedure, the assumptions and results of the Aspen simulation are discussed first. The most important assumptions of the LCA and the sensitivity analysis are then discussed before the used methods and the societal relevance of this work is examined.

6.1. Discussion of process simulation

The Aspen simulation of the filter was strongly influenced by the simplification that no interaction between the sludge and the water takes place. As a result, the energy consumption of the simulated process is 27% lower than the energy consumption described in the literature. This is due to the interstitial water, which is located within a sludge floc compound and requires more mechanical and therefore more electrical power to squeeze it out. In practice, this means that a filter press has to use more energy to achieve a higher solid content. Since in the Aspen simulation all water in the sludge is considered free water, this difference is not reflected in the energy consumption. Therefore, the filtration process of the business-as-usual scenario has the same energy consumption as the baseline scenario, although the solids content of 27% is significantly lower than the 35% solids content of the baseline scenario. As the assumption has such a big impact on the energy consumption, the literature value for filtration was used for the LCA.

In the drying process, another simplification is integrated in the Aspen model: for heating the feedstock, only the specific heat factor (Cp value) of water is taken into account, but not that of the sludge. Since the Cp value of sludge (1 J/kgK) is lower than that of water (4,2 J/kgK), this conservative approach results in 0,8% higher energy consumption compared to the thermodynamic optimum. In practice, this simplification is not significant since the energy consumption is multiplied by efficiency factors of 75 – 90% to determine the energy demand of the dryer. Even though Barry (2019) still uses an efficiency of 75%, the author has already anticipated that future technologies will increase the efficiency of the drying process (Barry et al., 2019). Since Barry's publication, the company Huber has installed a 90% efficient dryer in Innsbruck (Huber SE, 2023). Therefore, this study uses the 90% efficiency, which reduces the energy consumption of the dryer by over 300 kWh per ton char compared to a 75% efficiency.

Thanks to the extensive literature on the slow pyrolysis process (Jaramillo-Arango et al., 2016), this study does not need to investigate the effects of different reaction conditions but can build on the work of others. Knowing the performance at given reaction conditions, this study was able to investigate the energy consumption of the pyrolysis reaction in more detail. In this analysis, a new parameter was found to significantly affect the energy consumption of the reaction: the ash content of the feedstock. Since the ash is inert and does not participate in the reaction, a higher ash content in the feedstock leads to a lower energy consumption in the pyrolysis reaction. This influence has not been described in the literature so far and should be verified with experimental tests.

The combustion of the pyrolysis products simulated in Aspen provides information about the thermal energy released and the CO_2 produced. The amount of nitrogen oxides formed during combustion is limited to the nitrogen content of the feedstock. In practice, however, most nitrogen oxides are formed by the reaction of nitrogen present in the air, which is converted to nitrogen oxides at high combustion temperatures. Since Aspen does not calculate this part of the nitrogen oxides, the flue gas cleaning of a comparable EcoInvent process had to be used for the LCA. The selected process "incinerating sewage sludge" also emits substances that remain in the char or bio-oil. In order not to underestimate the environmental impact, a conservative approach was chosen for the selection of the emitted substances. Thus, an experimental analysis of the exhaust gases could lead to more accurate values and thus probably to lower emissions.

Although the simulated condenser did not result in perfect material separation between bio-oils and non-condensable gases, the heat balance of the condenser could be determined. This showed that the heat energy of the liquid outflow is sufficient to preheat the quench water. The energy consumption of the condenser therefore consists only of the water pump required for the quench water.

In order to simulate the process as realistically as possible, the energy consumption of the pumps for the process water and the compressor for the fresh air required for combustion was also modeled. In the baseline scenario, these together account for 34% of the electricity consumption, which shows the necessity of simulating these process supporting components.

6.2. Discussion of the LCA

The energy consumption of the different processes and the CO_2 emissions from the combustion of the pyrolysis products could be used directly for the LCA analysis. The linkage with the large LCA database EcoInvent completed the process with the necessary background processes.

A key factor in the LCA is the resolution of multifunctional processes. By using the excess heat for other processes that were not specified in this study but are part of the larger H2Steel value chain, it was possible not only to obtain a more realistic picture of actual emissions but also to avoid multifunctionality as the heat energy stayed in the system boundaries. In the co-production of bio-oil, multifunctionality can no longer be avoided. Besides system expansion, the chosen strategy, substitution or allocation were also possible approaches to solve multifunctionality. In the case of substitution, a bonus would have been attributed to the bio-oil scenario for the avoided emissions from petroleum production. These amount to only 0,4% of the business-as-usual scenario and would therefore have no significant impact. However, an economic allocation would have a larger impact on the results. Depending on the study, the price of bio-oil varies between 0,4 and 0,85 €/l, while the price of char varies between 0,07 and 2,5 €/kg (Campbell et al., 2018). This means that a large proportion of emissions would be attributed to bio-oil rather than to char, thus significantly reducing the environmental footprint of char production. Another way to gloss over the results is to exclude non-fossil CO_2 emissions from the climate change impact category. To account for the actual emissions of the process, fossil and non-fossil CO_2 were treated equally in the analysis and the system expansion approach was used for the multifunctional process.

In the LCA, the 3 technology scenarios were compared with the respective business-as-usual alternatives. Due to the extra energy required for the Torwash process, it was more environmentally damaging than the baseline scenario in all impact categories, and with the exception of climate change and freshwater eutrophication, also worse than the business-as-usual alternative. For this reason, the Torwash scenario was not considered for the further sensitivity analysis. Although the bio-oil scenario with resistant heater was also more environmentally damaging than the baseline scenario in all environmental categories, it has a higher dependency on the CO_2 intensity of the electricity grid. At the same time, the environmental impact of electricity production is strongly dependent on the national electricity mix which is currently undergoing a major transition. Therefore, its impact was intensively investigated in a sensitivity analysis.

Since the H2Steel project is funded by the EU and could be implemented in all EU member countries if successfully completed, the sensitivity analysis examined the influence of the national electricity mix for all EU member countries. Since electricity production mainly influences the impact category climate change, the CO_2eq difference between the baseline scenario and the bio-oil scenario was plotted on a map. This makes it possible to determine in which country which technology scenario has the lower environmental impact for climate change. If resistant heaters are used for the heat production, the bio-oil scenario emits less CO_2 in about half of the EU countries. However, if the much more efficient heat pumps are used for heat production, the bio-oil scenario emits less CO_2eq in all investigated countries and should therefore be the preferred technology scenario. This trend is further enhanced when the future impact of the technologies is analyzed.

By using the premise software and applying the superstructure approach, the integrated assessment models (IAMs) could be linked to the EcoInvent database, thus enabling the transition of electricity generation to impact not only foreground processes, but also background processes. Since less electricity is used in the baseline scenario compared to the bio-oil scenario with heat pump, the reduction in CO_2eq emissions here is 9%, less than half of the 20% achieved in the bio-oil scenario by 2030. The ongoing transition leads to a reduction in CO_2eq emissions by 2040, resulting in a total of 3.020 kg CO_2eq in the bio-oil scenario and to 3.390 kg CO_2eq per ton of char in the baseline scenario. For these results, the more conservative IAM was used, which corresponds to current national contributions and ultimately to a warming of 2.5° C by 2050. The more positive IAM (SSP2-PkBudg500), which is consistent with the Paris Agreement, would reduce GHG emissions by an additional 4% in the bio-oil scenario in 2040.

6.3. Discussion of the used methods

LCA is a core methodology of Industrial Ecology and can be used to provide process developers with an additional decision criterion besides cost, i.e. the environmental impact. The prospective LCA carried out in this work required many inputs that were not known to the process developers of the H2Steel project team at the early starting point of the project. Since those information were not yet described in detail in the literature, a process simulation had to be carried out. Even though the simulation performed in Aspen Plus showed certain deviations from the values described in the literature, it was still possible to obtain valuable information for processes not yet described in literature. Since Aspen allows a quick adjustment of the input variables, different scenarios could be investigated, and new process knowledge could be gained. In addition, details that are usually not described in the literature, such as the amount of air necessary for the combustion process and the amount of water necessary for the steam production, could be determined. Therefore, process simulation was of great value for the prospective LCA and thus contributed to answer the main research question.

In addition to the ecological impact of the technologies, the costs also play a major role in the decision-making process. Although an economic evaluation is outside the scope of this thesis, a superficial calculation of the operating costs was carried out. The baseline scenario consumes about 280 kWh of electricity per ton of char produced. At the current Italian electricity price of 0,15 € per kWh of electricity (Statista, 2022), this represents 42€. The approximate cost of the chemicals required for exhaust gas cleaning is 10€ per ton of char (Appendix 7.4). On the other hand, the disposal of sewage sludge costs 300 - 500€ per ton of wet sewage sludge (moisture content around 73%)(Jules van Lier, 2023b). Since 8,6 tons of wet sewage sludge (MC of 73%) are needed to produce 1 ton char, the disposal costs represent 2.500 – 4.300 €. It is very likely that this difference exceeds the unconsidered costs of the slow pyrolysis process like maintenance works, transport of the char to the steel plant or amortization of investment costs. Also, the co-production of bio-oil is economically profitable. One liter of bio-oil has an economic value between 0,4 and 0,85€ (Campbell et al., 2018). In the production of one ton char, 120 l of bio-oil can be obtained, which corresponds to an economic value of 50 - 100 €. This also significantly exceeds the cost for the energy required to provide heat, which comes to a price of 40 € per ton of char, if the Italian electricity price for small industries of 2021 is considered (Statista, 2022).

6.4. Discussion of the societal relevance

The question of the initial problem statement of how to make the steel industry more sustainable is answered in detail in Tanzer's report. It is described there that with consistent use of bioenergy and carbon capture and storage, even negative emissions in the steel industry are possible (Tanzer et al., 2020). The production of char analyzed in this report can become an important component of the bioenergy described in Tanzer's report. With the subsequent H2Steel "biomethane cracking" process, in which bio-methane is split into hydrogen and solid carbon, the value can be increased even further. The carbon deposits increase the carbon content of the char, making it a viable alternative to fossil coal. In addition to the upgraded char, the hydrogen produced in the process can also be used in the steel industry, contributing to its decarbonization.

The second initial problem relates to the end-of-life treatment of sewage sludge. Here, too, the technology analyzed shows great potential, as it can reduce the CO2eq emissions by up to 42% and also has lower impacts in all other environmental impact categories compared to the business-as-usual alternative, which involves the incineration of sewage sludge. In addition, the material utilization of sewage sludge proposed in the H2Steel project represents a higher hierarchy level than thermal utilization by incineration and should therefore be preferred after the European Directive 2018/851/EU.

The technology developed by H2Steel manages to transform the waste stream of one industry into the feedstock of another industry. This corresponds with the European Innovation Council goal to "Capturing cross sectorial coupling and system integration opportunities entirely based on (I) renewable sources and (II) non-toxic, non-critical raw materials" (EIC, 2022) and is thus of great societal value.

6.5. Conclusion

The objective of this report is to identify the most sustainable scenario to produce char from sewage sludge. For this purpose, the literature was first scoured to find the latest developments in the key processes. In the next step, these were simulated in the Aspen Plus software to obtain process-specific data on energy consumption and CO_2 emissions. In an LCA, these were combined with the necessary background processes to analyze the environmental impact of the entire value chain.

Based on the presented results, the main research question: "What is the most environmentally sustainable scenario to produce char from sewage sludge", can be clearly answered. Filtration should be carried out with a filter press, which requires 20% less energy than a centrifuge. For the drying step, a highly efficient two-chamber belt dryer should be used, as it consumes 15% less electricity than a conventional dryer. Due to the limitations of the data basis, it was not possible to finally decide whether the Torwash process reduces the environmental impact and should therefore be included in the process chain. If the missing process heat is replaced by a highly efficient heat pump, the GHG emissions in all EU countries are lower if the bio-oils are condensed than if the bio-oils are burned for heat production. This difference will become even greater in the future as electricity production relies less and less on CO_2 intensive fossil fuels. The phase-out of fossil fuels results in a reduction of the environmental impact in the two impact categories climate change and acidification. If a conservative integrated assessment model which leads to 2.5°C warming by 2050 is used as the basis for technological change, the emissions of the bio-oil scenario in the impact category climate change are 3.020 kg CO_2eq per ton of char, more than 9% lower than the baseline scenario and 42% lower than the business-as-usual alternative. In the impact category acidification, the bio-oil scenario performs 13% better than the baseline scenario and 22% better than the business-as-usual alternative. Although the ex-ante LCA does not predict a reduction in the other three impact categories (freshwater ecotoxicity, freshwater eutrophication and marine eutrophication), it can be assumed that technological progress in the wastewater treatment will also reduce the impact here.

Based on the results, the strong recommendation is expressed to produce char of sewage sludge according to the bio-oil scenario, with includes the latest drying and heat-pump technologies as well as the co-production of bio-oil. Since the end-of-life treatment of sewage sludge by slow pyrolysis and condensation of bio-oils is economically viable and also offers the possibility of decarbonizing the steel industry, this technology should be implemented on a large scale.

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7. Appendix

7.1. Aspen Inputs

In Table 1 the reactor types used in the Aspen model and the input parameters necessary for their simulation are shown.

Table 1 Assumptions for the specific processes

	Process details	Value	Unite
SP1-F1 Filter	Type: Solid Separator		
	Inlet solid concentration	4	%
	Inlet temperature	20	°C
	Inlet composition defined in Table 2		
	Outlet solid concentration (chamber filter press)	35	%
	Particle size distribution defined in Table 3		
SP1-DR1 Dryer	Type: Continuous, Shortcut		
	Temperature	101	°C
SP1-R1 Slow Pyrolysis Reactor	Type: RYield		
	Temperature	400	°C
	Char yield (composition in Table 4)	42	%
	Gases yield (composition in Table 5)	32	%
	Bio-fuels yield (composition in Table 6)	26	%
SP1-SP1 Cyclone	Type: Cyclone		
	Fraction solid to solid outlet	0,99	
	Fraction vapor to vapor outlet	0,99	
SP1-FU1 Combustion Chamber	Type: RStoic		
	Combustion reaction with NO_x combustion products		
	Air fuel ratio	10 to 1	
	Air pressure	1,3	bar
SP1-E1 Heat Exchanger	Type: Heater		
	Outlet Temperature	500	°C
SP-E2 Heat Exchanger	Type: Heater		
	Outlet Temperature	150	°C
SP-E3a Heat Exchanger	Type: Heater		
	Outlet Temperature	200	°C
SP-E3b Heat Exchanger	Type: Heater		
	Outlet Temperature	40	°C
SP-E4 Heat Exchanger	Type: Heater		
	Outlet Temperature	60	°C

7.1.1. Composition of the investigated digested sewage sludge

Table 2 Proximate and ultimate analysis of the sewage sludge as well as the composition of the resulting ash (based on Jaramillo-Arrango et al., (2016))

		Analytical method	Dry basis (d.b)
Proximate analysis (% in mass)	Moisture	ASTM D 3173	-
	Ash	ASTM D 3174	67,5
	Volatiles	ASTM D 3175	19,01
	Fixed Carbon	ASTM D 3172	3,49
Ultimate Analysis (% in mass)	Carbon	Elemental analyses using a Carlo Erba 1108.	12,79
	Hydrogen	Elemental analyses using a Carlo Erba 1108.	1,74
	Oxygen	by difference	16,22
	Nitrogen	Elemental analyses using a Carlo Erba 1108.	1,2
	Sulfur	ASTM D 4239	0,55
	Ash		67,5
Composition of ash (% in mass)	SiO2	CEN/TS 15290.	48,92
	Al2O3	CEN/TS 15290.	21,25
	Fe2O3	CEN/TS 15290.	7,42
	CaO	CEN/TS 15290.	6,23
	P2O5	CEN/TS 15290.	3,89
	MgO	CEN/TS 15290.	2
	K2O	CEN/TS 15290.	1,43
	Na2O	CEN/TS 15290.	1,18
	TiO2	CEN/TS 15290.	1,18
	MnO2	CEN/TS 15290.	0,16

7.1.2. Particle size distribution of the investigated sewage sludge

Table 3 Particle size distribution of sewage sludge based on Funke et al., (2018))

Lower limit [µm]	**Upper limit [µm]**	**Weight fraction [%]**
0,45	6,9	0,1
6,9	30,5	0,4
30,5	292,2	0,5

7.1.3. Composition of the Char

Table 4 Elemental analysis of char produced at 400°C in a fluidized bed (% in mass) (based on Jaramillo-Arrango et al., (2016))

Elemental analysis of char	**Amount**
C	12,90%
H	0,50%
N	0,21%
S	0,52%
Ash	85,87%

7.1.4. Composition of Bio-oils

Table 5 shows the composition of bio-oil produced in the experiments of Jaramillo-Arango et al., (2016). The column original values list the experimental values of all compounds above 2%. To replace the number of compounds below 2% in the simulation, the experimental values were tuned.

Table 5 Bio-oil composition based on (based on Jaramillo-Arrango et al., (2016))

Compound	Original value	Tuned value
Acetronitrile	2,42	2,96
Pyridine	2,43	2,97
Styrene	3,83	4,68
2-Cyclopenten-1-one, 2-methyl-	1,81	2,21
Acetic acid	4,77	5,83
1-Hexanol, 2-ethyl-	6,82	8,33
1-Ethylcyclopentene	8,92	10,90
Acetamide	4,28	5,23
Benzyl nitrile	2,38	2,91
Phenol, 2-methyl-	2,37	2,89
Phenol	10,89	13,30
2-pyrrolidinone	4,83	5,90
Phenol, 4-methyl-	12,87	15,72
Phenol, 4-ethyl-	3,74	4,57
Indole	5,12	6,25
1,2,4-Triazine-3,5(2H,4H)-dione	2,42	2,96
1H-Imidazole, 2-methyl-	2,01	2,41
Total	**81,87**	**100**

7.1.5. Composition of pyrolysis gases

Table 6 Composition of non-condensable gases at 400°C (based on Jaramillo-Arrango et al., (2016))

Gas	Amount
CO_2	69%
CO	18%
CH_4	3%
H_2	10%

7.2. Inventory Table

Table 7 Inventory table, biosphere flows are displayed in cursive

Process / Sub-process	Amount	Unite
Baseline filter cake 35% SS (filter press)	6.645	kg
treatment of wastewater from anaerobic digestion	-58	m^3
acrylic acid production (EcoInvent)	14	kg
electricity	283	kWh
wastewater from anaerobic digestion	65	m^3
Baseline dried filter cake	2.326	kg
Baseline process heat	3.539	kWh
Baseline filter cake (filter press)	6.645	kg
water (air, non-urban air or from high stacks)	*3.813*	*kg*
Baseline char produced by slow pyrolysis	1.000	kg
bio-oil	-598	kg
non condensable gases	-818	kg
Baseline process heat	639	kWh
Baseline dried filter cake	2.326	kg
excess heat	564	kWh
Baseline process heat	4.742	kWh
bio-oil	598	kg
non condensable gases	818	kg
Baseline exhaust gas purification	92.700	m^3
Compressed air	11.610	kg
Process water (3 bar)	11	m^3
Compressed air	11.610	kg
carbon dioxide, non fossil (air - urban air close to ground)	*1.827*	*kg*
Compressed air	1	kg
electricity	6,05E-03	kWh
fresh air	1	kg
Exhaust gas purification	1	m^3
quicklime, milled, packed (EcoInvent)	7,77E-06	kg
hydrochloric acid, without water, in 30% solution state (EcoInvent)	1,25E-06	kg
iron (III) chloride, without water, in 40% solution state (EcoInvent)	5,04E-07	kg
sodium hydroxide, without water, in 50% solution state (EcoInvent)	1,68E-04	kg
ammonia, anhydrous, liquid (EcoInvent)	5,58E-05	kg
Nitrate (water - surface water)	1,40E-05	*kg*
Nitrogen oxides (air - urban air close to ground)	4,93E-05	*kg*
Dinitrogen monoxide (air - urban air close to ground)	1,64E-05	*kg*

Sulfur dioxide (air - urban air close to ground)	5,13E-06	*kg*
Nitrate (water - ground-, long-term)	3,95E-05	*kg*
Sulfate (water - surface water)	2,45E-04	*kg*
Ammonia (air - urban air close to ground)	1,56E-07	*kg*
Process water (3 bar)	1	m^3
electricity	2	kWh
heat produced by a resistant heater	1	kWh
electricity	1	kWh
heat produced by heat pump	4	kWh
electricity	1	kWh
Torwash treated digest	14	m^3
wastewater from anaerobic digestion	14	m^3
Torwash process heat	2.513	kWh
Torwash filter cake 50% SS (filter press)	4.651	kg
treatment of wastewater from anaerobic digestion	-60	m^3
acrylic acid production (EcoInvent)	14	kg
electricity	283	kWh
wastewater from anaerobic digestion	65	m^3
Torwash dryed filter cake	2.326	kg
process heat	1.667	kWh
Baseline filter cake (filter press)	4.651	kg
Water (air, non-urban air or from high stacks)	*2.325*	*kg*
Torwash char produced by slow pyrolysis	1.000	kg
bio-oil	-598	kg
non condensable gases	-818	kg
Torwash process heat	639	kWh
Baseline dryed filter cake	2.326	kg
excess heat	564	kWh
Torwash process heat	5.383	kWh
bio-oil	598	kg
non condensable gases	818	kg
Exhaust gas purification	92.700	m^3
Compressed air	11.610	kg
Heat produced by heat pump	2.753	kWh
Process water (3 bar)	11	m^3
carbon dioxide, non fossil (air - urban air close to ground)	*1.827*	*kg*

Bio-Oil dryed filter cake	2.326	kg
Bio-Oil process heat	3.539	kWh
Baseline filter cake (filter press)	6.645	kg
water (air, non-urban air or from high stacks)	*3.813*	*kg*
Bio-Oil char produced by slow pyrolysis	1.000	kg
bio-oil	-598	kg
non condensable gases	-818	kg
Bio-Oil process heat	639	kWh
Baseline dryed filter cake	2.326	kg
Baseline excess heat	564	kWh
Bio-Oil condensation of bio-oil	598	kg
bio-oil	598	kg
Process water (3 bar)	3	m³
Bio-Oil process heat	4.742	kWh
non condensable gases	818	kg
Exhaust gas purification	53.000	m³
Bio-Oil compressed air	4.693	kg
Process water (3 bar)	5	m³
Heat produced by heat pump	4.475	kWh
carbon dioxide, non fossil (air - urban air close to ground)	*1.644*	*kg*
Business-as-usual filtration	8.614	kg
treatment of wastewater from anaerobic digestion	-58	m³
Acrylic Acid	14	kg
electricity	283	kWh
wastewater from anaerobic digestion	65	m³
Business-as-usual sludge incineration		
treatment of raw sewage sludge, municipal incineration (EcoInvent)	-8.614	kg
Business-as-usual sludge filtration	8.614	kg
Business-as-usual coal provision	313	kg
market for hard coal (EcoInvent)	313	kg
Business-as-usual petroleum provision	109	kg
market for petroleum (EcoInvent)	109	kg

7.3. Process contribution analysis

The following graphs represent the process contributions for all impact categories. The process heat for the Torwash and bio-oil scenario is provided by a heat pump.

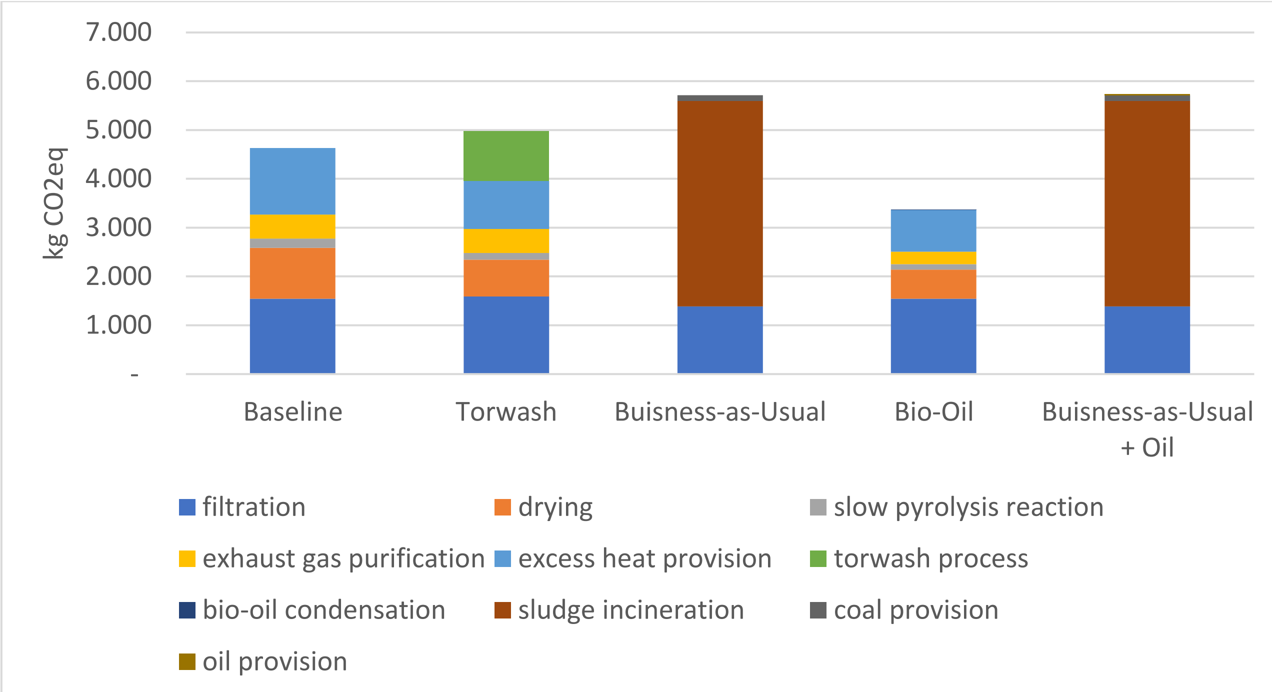

Figure 24 Process Contributions for the impact category climate change

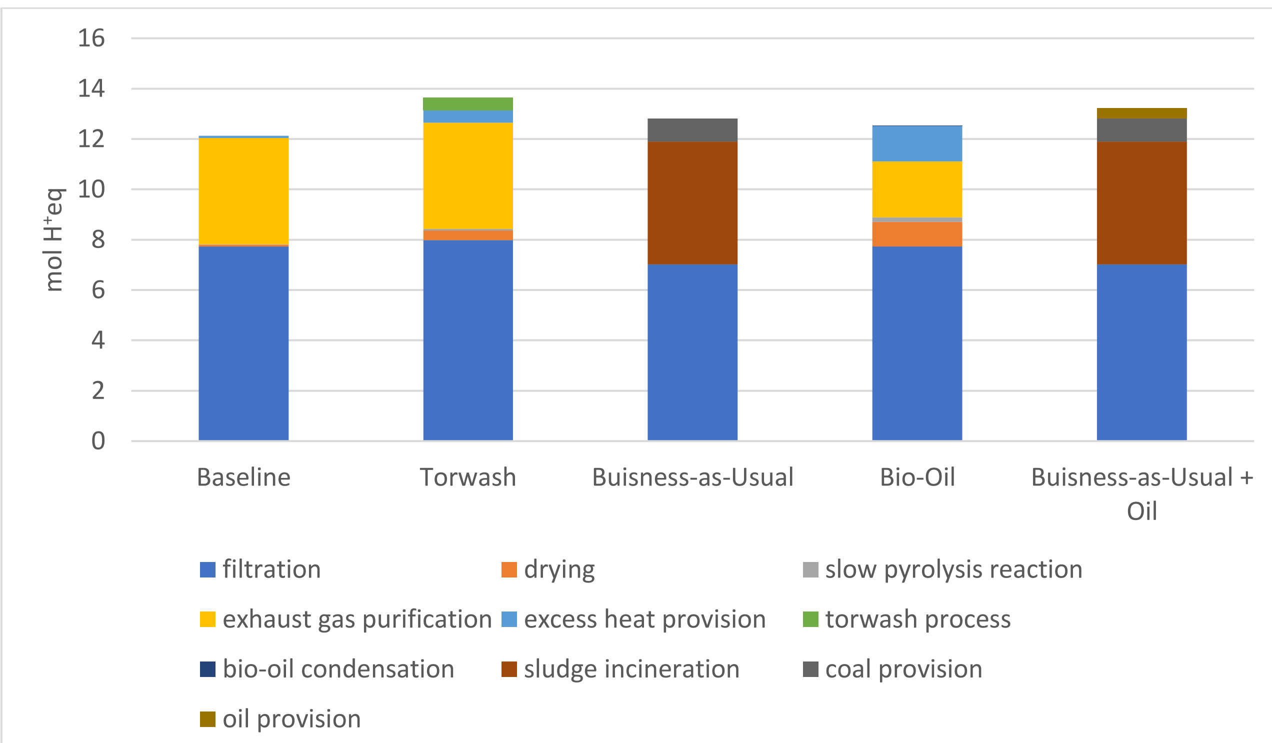

Figure 25 Process contributions for the impact category acidification

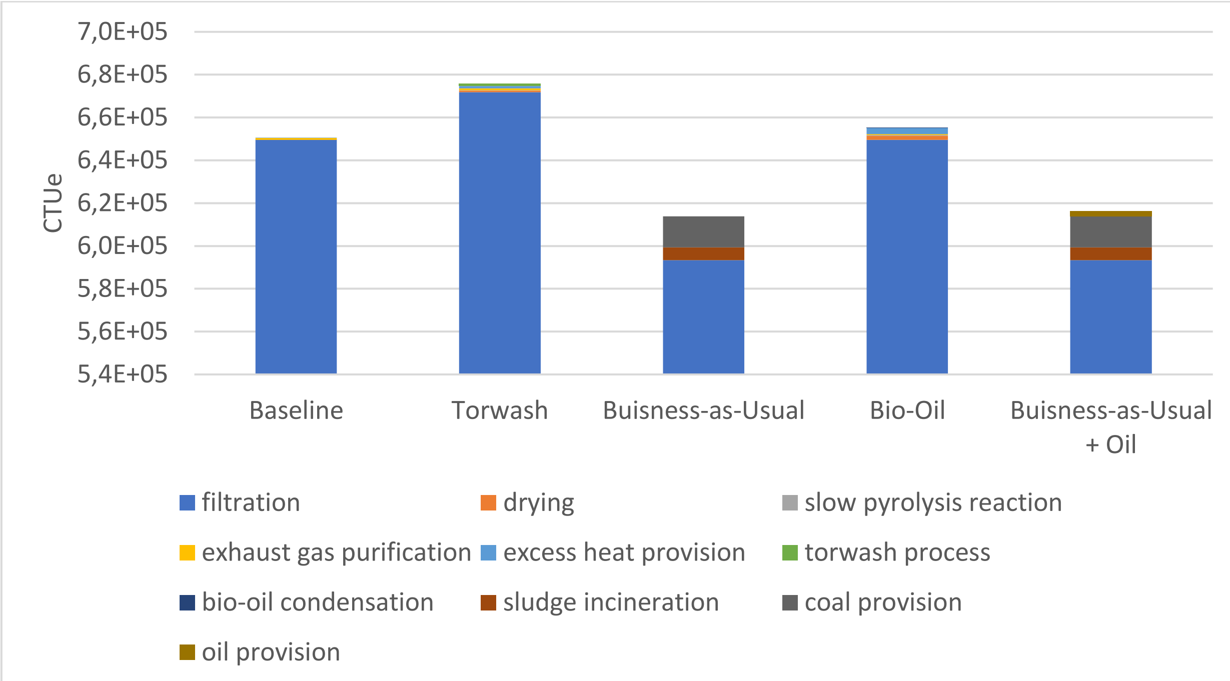

Figure 26 Process contributions for the impact category freshwater ecotoxicity

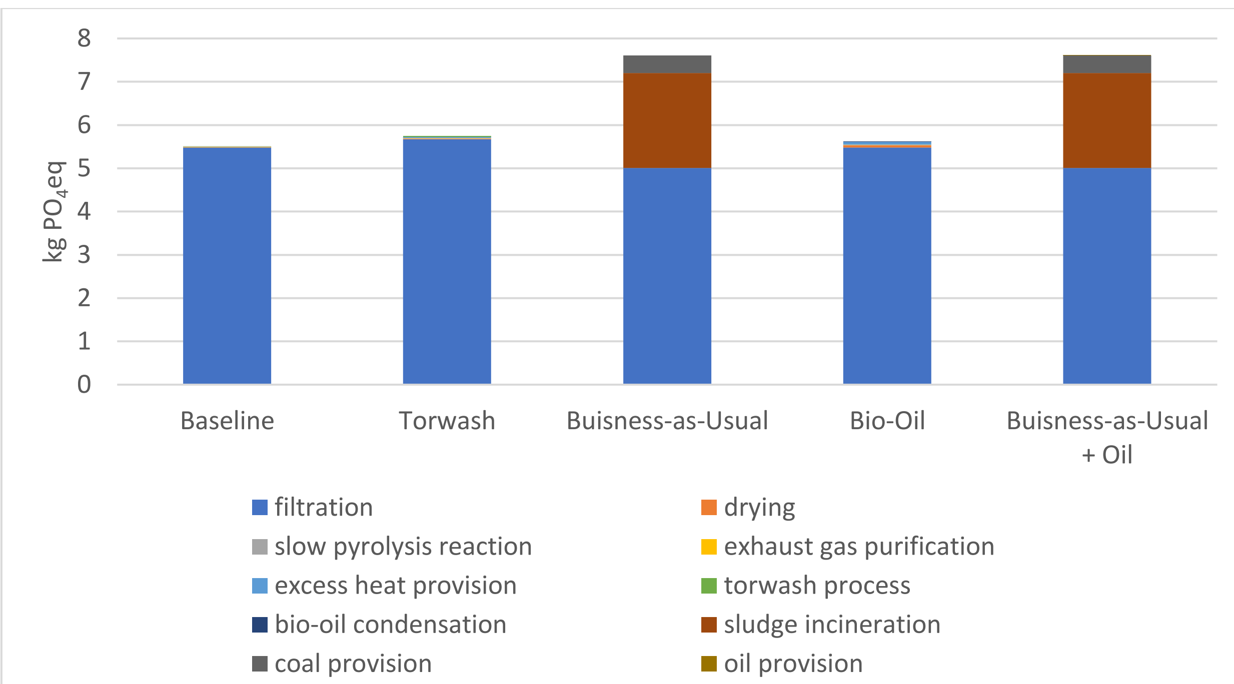

Figure 27 Process contributions for the impact category freshwater eutrophication

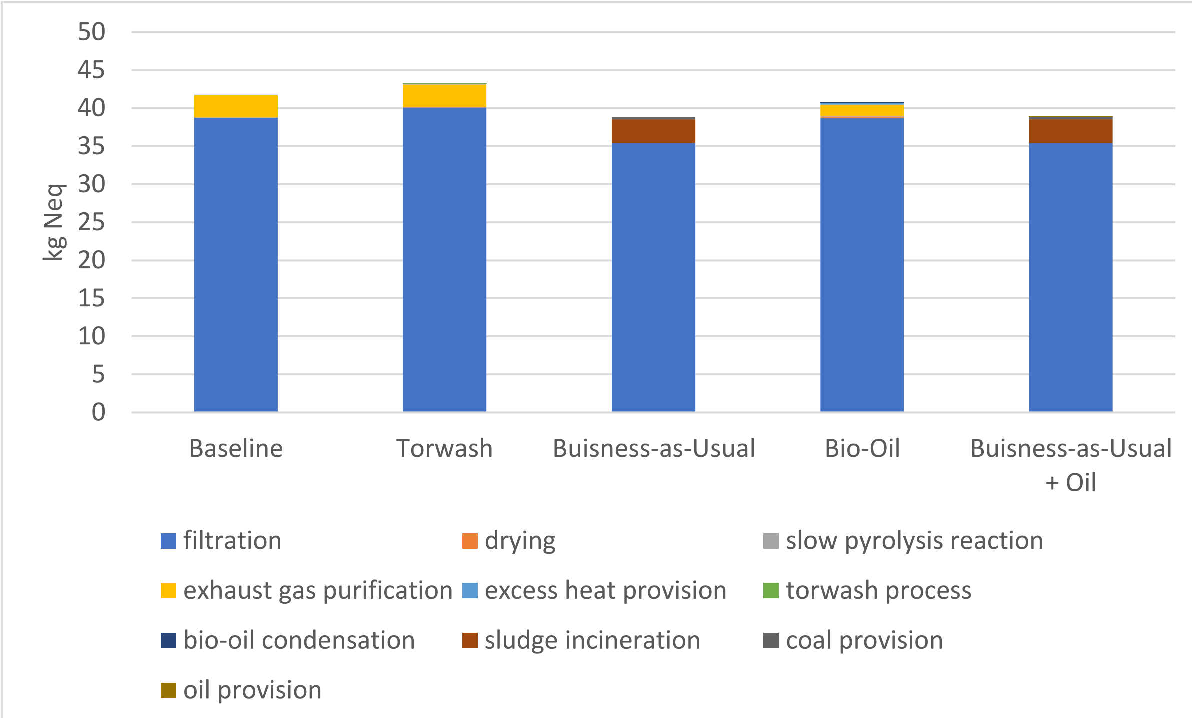

Figure 28 Process contributions for the impact category marine eutrophication

7.4. Rough estimate of the costs for exhaust gas cleaning

Table 8 Estimation of the costs for exhaust gas cleaning

	€/ton char	**€/kg**	**kg/ton char**	**Source**
Quicklime	0,07	0,10	0,72	(Alibaba.com, 2023a)
Hydrochloric acid 30% solution	1,58	13,65	0,12	(Lab Alley, 2023)
Iron (III) cloride	1,96	41,83	0,05	(Avantor, 2023)
Sodium Hydroxide	6,24	0,40	15,59	(Alibaba.com, 2023c)
ammonia liquid	0,32	0,06	5,17	(Alibaba.com, 2023b)
Total	**10,16**			