

This is the preprint version of the contribution published as:

Reißmann, D., Thrän, D., Bezama, A. (2018):

[Hydrothermal processes as treatment paths for biogenic residues in Germany: A review of the technology, sustainability and legal aspects](#)

J. Clean Prod. **172** , 239 – 252

The publisher's version is available at:

<http://dx.doi.org/10.1016/j.jclepro.2017.10.151>

1 Total words: 11.472

2 Hydrothermal processes as treatment paths for
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11 KEYWORDS: Hydrothermal Processes; Review; Biogenic Residues; Germany

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Abstract

A considerable part of especially wet and sludgy biogenic residues is currently not in material or energetic usage in Germany. Therefore, a key issue for current research is to identify which technologies are most suitable at mobilizing these wet and sludgy materials. Hydrothermal Processes (HTP) appear to be promising treatment options for moist substrates because they require a high water content of 70% to 90% for optimal processing. This review provides information on the state of the art and knowledge on HTP, and attempts to determine how suitable these processes are for mobilizing biogenic residues in Germany. We identified technological, economic, environmental and legal potentials and barriers of HTP using a modified content-analysis. About 120 relevant references were identified and analyzed using a structured sampling scheme. The results show considerable advantages of HTP for utilizing wet and sludgy biogenic residues in contrast to comparable biomass treatment processes. Especially, their high process energy-efficiency and low Global Warming Potential from a life cycle perspective. Nevertheless, technological, economic, environmental and legal barriers (e.g. missing data and knowledge on process kinetics; missing legal standards) must be taken into consideration. Finally, research needs are illustrated that must be fulfilled through structured and target-oriented research.

1. Introduction

Biogenic residues from industrial, commercial and municipal activities are valuable resources. Residues like liquid manure, straw, wood residues from the forestry industry, industrial wood residues, demolition wood, kitchen and garden waste, sewage sludge, and municipal solid waste, can be utilized in a value-enhancing way through appropriate technological applications (Leible et al., 2003; Tröger et al., 2013). The German Government already fosters the material and energetic utilization of biogenic residues by several programs, initiatives and legal regulations aiming to increase the resource efficiency of process chains (BMUB, 2016). Due to disposal regulations specified through the German Law on Closed Cycle Management and Waste (KrWG, 2012), most industrial residues like plant oils and animal fats as well as municipal waste streams such as food and bio-waste are already being utilized (Brosowski et al., 2016). Regarding the technical potential - describing the part of all physically existing biogenic residues for a certain region and time that is applicable under consideration of availability, environmental barriers (e.g. erosion), technical feasibility, competing uses and legal requirements (Brosowski et al., 2016) - approximately 30% is currently used to produce materials (e.g. compost; fertilizers; cosmetics; pharmaceuticals; bio-plastics) through mainly chemical and physical conversion processes (cf. Thrän and Bezama, 2017; Spiridon et al., 2016; Türk, 2014). A further 27% of the technical potential is energetically used to produce electricity, fuels and heat through thermochemical and biochemical processes (cf. Long and Karp, 2013; Okoro et al., 2017). However, in addition to the substrates that are already tied to material and energetic treatment paths, a technical potential of around 30 million tons of biogenic residues are currently not being used in Germany (Brosowski et al., 2016). Wood residues, cereal straw, animal excreta and sewage sludge are particularly often not in energetic or material use in Germany. Moreover, many biogenic residues used in thermal processes are not

suitable because their heating value is under 11 MJ/kg (Brosowski et al., 2016). In addition, some treatment paths for biogenic residues have the potential to increase efficiency through process cascades, i.e. the expansion of existing process chains through material recovery and recycling (Bezama, 2016; Thonemann and Schumann, 2016). With this in mind, the question arises as to whether and how this unused potential can be mobilized, and which processes are most suitable for this purpose.

Hydrothermal processes (HTP) appear to be a promising technology platform for processing wet and sludgy biogenic residues. These technologies use water as their main process medium to convert biomass into materials and fuels at high pressures and temperatures. Because a very moist environment is needed to ensure that the process runs effectively, less energy and thus costs are required in contrast to conventional treatment paths because process steps like substrate thickening and drying are not needed anymore. This makes HTP interesting from an economic and environmental point of view (Schindler, 2015). Thus, HTP seem to be a suitable way to mobilize the wet and sludgy part of the unused biogenic residues in Germany. However, the novelty of the technology platform is associated with uncertainties and barriers for stakeholders (e.g. investment decisions, development of legal standards, funding decisions etc.). Hence, this review aims to contextualize HTP based on technological, economic, environmental and legal criteria.

2. Structure of the review and methods

This review follows the sequence illustrated in Figure 1. The process is oriented on a modified content-analysis with the aim to provide new insights and enhance the understandability of certain issues through a structured procedure (cf. Moldavska and Melo, 2017).

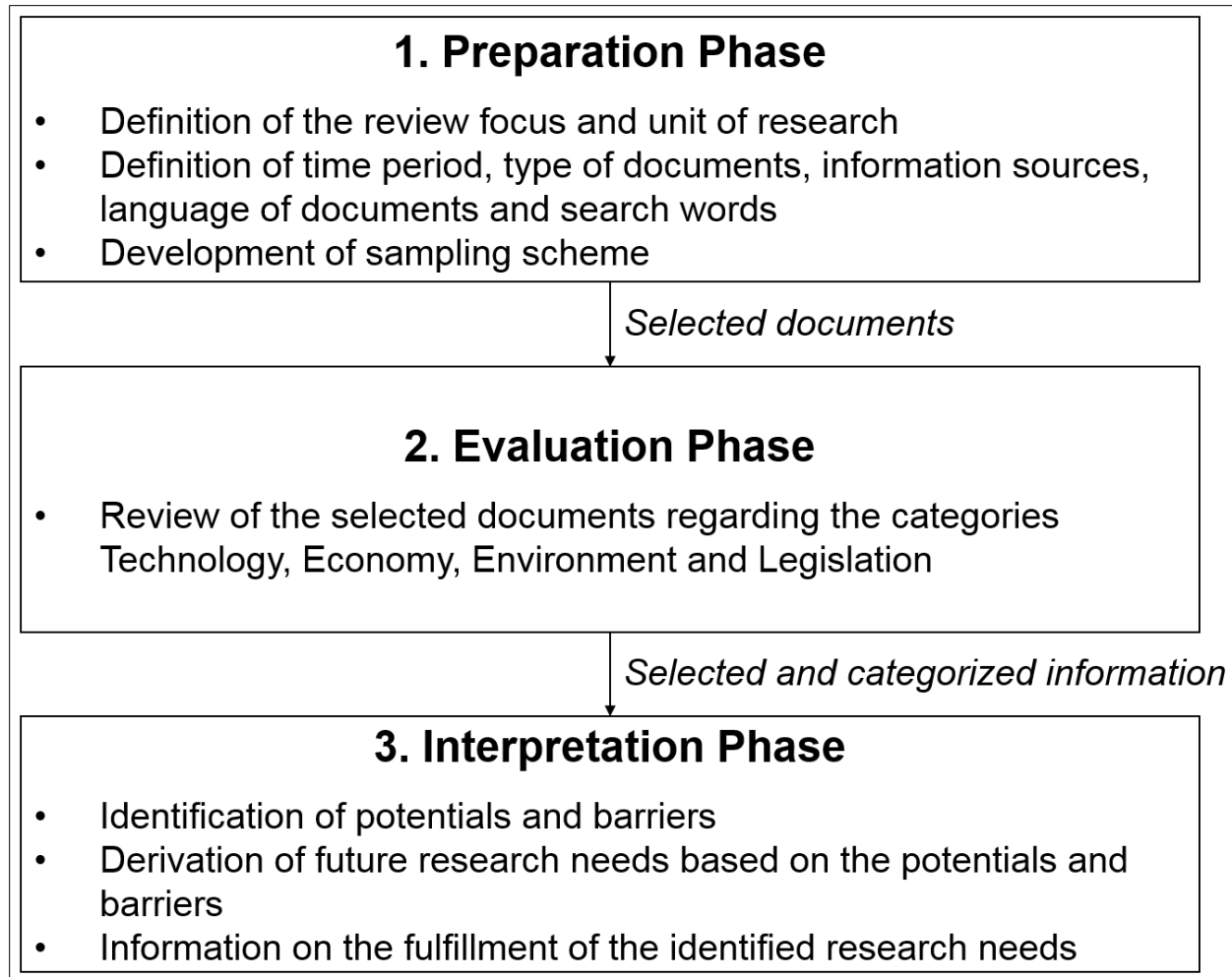


Figure 1. Sequence of the Review (adapted from Moldavska and Melo 2017)

Step 1: Preparation Phase

First, the review focus was defined according to the study purpose that is to evaluate the extent to which HTP represent a viable option for processing currently unused biogenic residues in Germany. Thus, the central focus was set to identify technological, economic, environmental and legal potentials and barriers of HTP, to derivate corresponding future research needs and to provide information on how to fulfill the research gaps. Based on the review focus, the unit of research was defined as scientific and practical information on the technological, economic, environmental

and legal potentials and barriers of Hydrothermal Processes as options for treating biogenic residues in Germany.

Second, a sampling focus including the definition of the time period, type of documents, information sources and document languages must be defined. Because the research on Hydrothermal Processes has gained rising attention since 2000, the period of consideration was set from 2000 to 2017. A large range of different document types was included into the review. Particularly, scientific articles and textbooks, presentations on scientific conferences, conference proceedings, technical reports, legislative texts and websites written in both German and English. The reason for the selection of these document types is that current research on HTP includes much applied-oriented research that is often published via technical reports. Next to this, most recent results are often presented on conferences or websites before they are published in scientific journals or textbooks. Thus, these types of documents should be considered next to scientific articles and textbooks. The information sources used were Google, Google Scholar, Science Direct and Scopus.

Third, to identify documents that are most relevant considering the review focus, we used a sampling scheme (Fig. 2). For every process step of HTP it was determined which information about the aspects under consideration (Technology; Economy; Environment; Legislation) was needed to fulfill the review purpose and thus the defined focus. Based on suggestions of Thrän et al. (2013) the most relevant keywords for each process step and aspect were identified accordingly.

Process Chain of HTP	Keywords on Technology	Keywords on Economy	Keywords on Environment	Keywords on Legal status
Feedstock and collection	<ul style="list-style-type: none"> suitable substrates biomass potential 	<ul style="list-style-type: none"> feedstock supply costs 	<ul style="list-style-type: none"> Life Cycle Assessment Life Cycle Performance LCA 	<ul style="list-style-type: none"> legal status of feedstock supply
Preparation/ Transport/ Storage	<ul style="list-style-type: none"> pre-treatment 			
Conversion & Refinement	<ul style="list-style-type: none"> process parameter process design 	<ul style="list-style-type: none"> investment costs operating costs 		<ul style="list-style-type: none"> process and plant standards
Distribution of products	<ul style="list-style-type: none"> products by-products process water product usage 	<ul style="list-style-type: none"> distribution costs transport costs 		<ul style="list-style-type: none"> product quality standards
Product Usage		<ul style="list-style-type: none"> sales 		<ul style="list-style-type: none"> product authorization

Figure 2. Sampling scheme to systematically identify the most relevant keywords for document research

The keywords shown in the boxes of the sampling scheme were used in connection with search words for HTP particularly “Hydrothermal Processes”, “Hydrothermal Carbonization”, “Hydrothermal Liquefaction” and “Hydrothermal Gasification”. The following bullet points clarify the search queries:

- “Hydrothermal Processes AND keyword” (e.g. “Hydrothermal Processes sales”),
- “Hydrothermal Carbonization AND keyword” (e.g. “Hydrothermal Carbonization products”),

- “Hydrothermal Liquefaction AND keyword” (e.g. “Hydrothermal Liquefaction feedstock supply costs”),
- “Hydrothermal Gasification AND keyword” (e.g. “Hydrothermal Gasification by-products”).

The above mentioned search words for HTP were also used without keywords from Figure 2 to identify more general documents on HTP. To reduce the risk that the search strategy applied could possible exclude relevant documents, an additional test research with more detailed keywords was applied. The words used for this were biogenic residues, municipal waste, sewage sludge and animal excreta. For the test search queries the mentioned words were also connected to the search words for HTP (cf. above mentioned bullet points). In result, the authors claim that the search strategy includes the most relevant documents because also through the test research mostly these documents were identified.

Step 2: Evaluation Phase

Through the keyword research about 120 relevant references were identified and analyzed, whereby not all of them are cited in this article because some information were part of various documents. Every document was carefully reviewed according to the search focus (i.e. the used keyword) and the underlying category (Technology; Economy; Environment; Legislation).

Step 3: Interpretation Phase

Based on the results of the review process, potentials and barriers to HTP for mobilizing the unused technical potential in Germany were identified and interpreted. Finally, future research needs and suggestion to fulfill these needs were derived.

3. Results

3.1. Technological issues of Hydrothermal Processes

3.1.1. Suitable feedstock, feedstock pretreatment and biomass potential for HTP in Germany

The water content is a key parameter for an efficient hydrothermal processing (Greve et al., 2014). An organic dry matter content of less than 30% is generally recommended (Greve et al., 2014; Libra et al., 2011; Ramke et al., 2012). However, the dry matter content of the substrate is the most important parameter for optimizing the desired product output per hour and invested monetary unit because the production rate of the desired output (coal, oil, gas) is proportional to the amount of biomass feed in (Vogel, 2016). Based on this, the suitable organic dry matter content should range between 10% and 30%.

To reach high product mass and energy yields, lignocellulose residues (e.g. corn stalk and dough residues) are very suitable for all HTP types (Kong et al., 2008; Libra et al., 2011; Oliveira et al., 2013; Xiao et al., 2012) whereby algae is the most suitable input for Hydrothermal Liquefaction (HTL – described in section 3.1.2.) (Zhang et al., 2015; Zhu et al., 2013). Generally, no expensive pretreatment is necessary when using the mentioned substrates in HTP. The only exception is that relatively solid substrates (e.g. stalks) must be sufficiently shredded into smaller particles to ensure uninterrupted pumping (Hoffmann, 2014). Based on the mentioned requirements, technical feasibility, structural conditions, ecological issues and social priorities, Brosowski (2015) calculated that Germany has a technical biomass potential for HTP of 16.8 million tons of dry matter. This includes 9.1 million tons of animal excreta, 5.7 million tons of sewage sludge and 2.0 million tons of stalk landscaping materials (Brosowski, 2015).

3.1.2. Parameters and process designs that influence the process

Different types of hydrothermal processes occur depending on pressure, temperature and residence time which is why these reaction parameters are crucial (Greve et al., 2014; Kruse et al., 2013; Peterson et al., 2008). Table 1 shows the typical ranges of these parameters for the main types of HTP: Hydrothermal Carbonization (HTC), Hydrothermal Liquefaction (HTL) and Hydrothermal Gasification (HTG), with its sub-reactions catalytic/low-temperature (subcritical conditions and addition of heterogeneous catalysts) and non-catalytic/high-temperature (supercritical conditions with addition of homogenous catalysts) processes (Elliott, 2008). The parameters are compared to anaerobic digestion as reference biomass conversion process.

Table 1. Typical temperatures, pressures and residence times for the main types of HTP (Data from (1) Boukis et al., 2003; (2) Kruse et al., 2013; (3) Peterson et al., 2008; (3) SEAI, 2016; (4) Vogel, 2016)

HTP type	Temperature range	Pressure range	Typical residence time range
HTC – Hydrothermal Carbonization	160-250 °C (2)	10-30 bars (2)	1-72 hours (4)
HTL – Hydrothermal Liquefaction	180-400 °C (2)	40-200 bars (3)	10-240 minutes (1)
HTG – Hydrothermal Gasification			
Catalytic/low-temperature	350-450 °C (4)	230-400 bars (3)	< 10 minutes (4)
Non-catalytic/high-temperature	> 500 °C (4)	230-400 bars (3)	< 10 minutes (4)
<i>Reference process: Anaerobic Digestion</i>	32-65 °C (3)	ambient pressure (3)	35-80 days (3)

Even though much higher temperatures are needed for HTP, the reactions are considerably faster than for the anaerobic digestion process. In addition to process parameters, the catalyst (Guo et al., 2013; Katarzyna et al., 2016; Kong et al., 2008), heating velocity (Katarzyna et al., 2016), solvent

(Xiao and Guo, 2006), substrate solid ratio (Dandamudi et al., 2016) and pH value of the feed (Funke, 2012) have a significant impact on the efficiency of the process and the characteristics of the products. Several studies mention a substantial catalytic effect of potassium chloride, citric acid (HTC), alkali carbonate, alkali hydroxide (HTL, HTC) and nickel (HTG) on the processing efficiency (Guo et al., 2013; Klemm et al., 2012; Kong et al., 2008). An optimized calibration of these parameters is recommended in order to ensure a high-quality product (e.g. high calorific value, low pollution level, high nutrient content) and an efficient process. Table 2 lists some calibration examples for temperature, pressure and residence time of specific process designs.

Table 2. Examples for the optimal calibration of temperature, pressure and residence for HTC, HTL and HTG

Process example	Temperature (°C)	Pressure (bar)	Residence time (sec)	References
Batch HTC with fermentation residues aimed at high nutrient contents in HTC-char	220	2	14400- 28800	Brookman et al., 2016
Batch HTL with waste furniture sawdust aimed at maximum bio-oil yield	280	10	900	Jindal et al., 2016
Continuous HTG with glucose in sub- and supercritical water aimed at high product gas yields	> 480	340	4	Klingler and Vogler, 2010

It should be noted that the suggestions mentioned in Table 2 are only valid for the specific process example under consideration and general recommendations have yet to be developed. To get a general impression of the efficiency of typical hydrothermal processes, Table 3 shows process efficiency ranges for HTC, HTL and HTG compared to thermochemical and biochemical biomass conversion processes. Here, process efficiency is based on the yield of the desired product in relation to the total dry matter feed in.

Table 3. Process efficiencies of HTP types compared to thermochemical and biochemical biomass conversion processes

Conversion type	Process	Process efficiency (%)	References
Biomass to coal	Slow pyrolysis (pyrolysis coke)	35	Ronsse et al., 2013
	Torrefaction	75	Ronsse et al., 2013
	HTC	70-90	Klemm et al., 2012
Biomass to liquid	Flash pyrolysis (pyrolysis oil)	65-75	Klemm et al., 2012
	HTL	70-86	Klemm et al., 2012
Biomass to gas	Gasification	54-58	Duret et al., 2005
	Anaerobic digestion	25-71	Weiland, 2010; Yoshida et al., 2003
	HTG	68-85	Klemm et al., 2012

Currently, the most common types of HTP processing systems are batch reactors and continuous-flow operating systems, whereby multi-batch systems are also used (cf. Badoux, 2011). Commonly used reactors are stirring tanks, barrels and tube reactors. Most plants operate as demonstration or pilot plants. The sewage sludge-based HTC process “SlurryCarb” (GlobalWaterAdvisors, 2017) is the most advanced type of HTP so far with the largest plant (located in Rialto, California, U.S.) converting about 180,000 tons of biomass fresh matter into HTC-coal per year (Bolin et al., 2007).

3.1.3. Products, product use and by-products

All of the HTP types produce solid, liquid and gaseous outputs whereby there is usually one intended output depending on the type of process used. The desired output of HTC is solid hydro-coal/HTC coal and bio-char. It can be used as a fuel (HTC coal), fertilizer and soil conditioner (bio-char). The liquid bio-crude or HTL oil is the main product of HTL which can be used as a bio-fuel and as a substitute for crude oil in chemical products like cosmetics. HTG mainly produces platform chemicals and bio-fuels based on a mix of hydrogen and methane (HTG product gas) (Kruse et al., 2013). Most HTP products are used for energetic purposes, e.g. as substitutes to

lignite, crude oil or natural gas (Vogel, 2016). Thus, the calorific value of the products is crucial. The following illustration shows typical ranges (minimum to maximum) of calorific values of HTC coal and HTL oil compared to conventional fuels. Because there is no robust data for HTG product gases, they are not included in the graph.

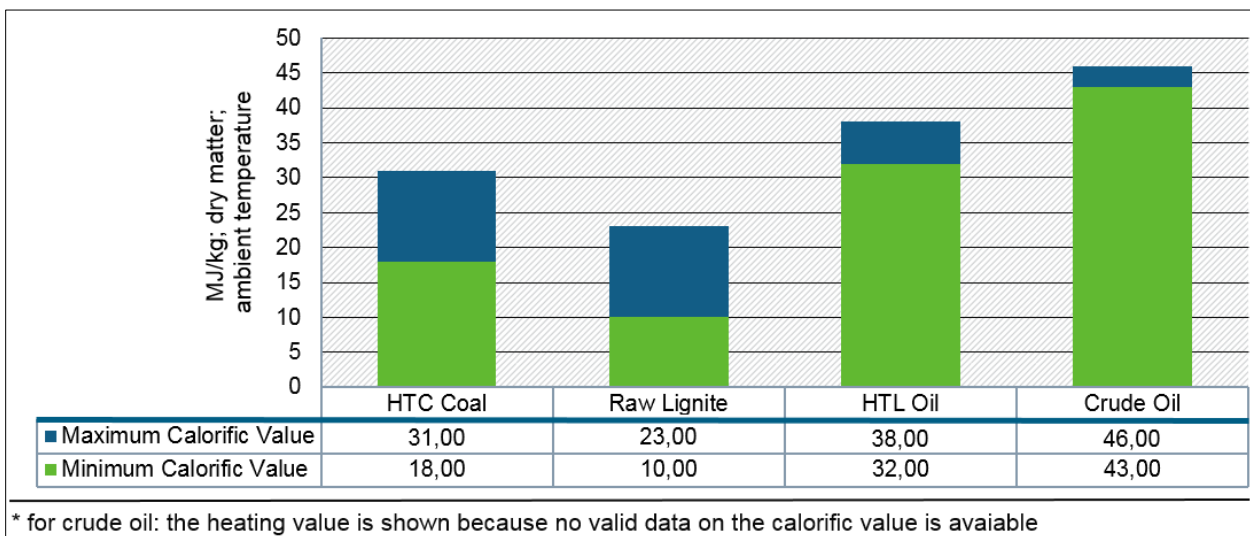


Figure 3. Maximum and minimum calorific values of HTC coal and HTL oil compared to raw lignite and crude oil in MJ/kg. (Data from Cerbe et al., 2008; GRENOL GmbH, 2012; Herrmann and Weber, 2011; Ramke et al., 2009; Vogel, 2016)

Most current applications refine the raw HTL oil afterwards through up-grading processes. This attains a higher quality which is comparable to conventional fuel. For example, HTL oil achieves a calorific value of 46.86 MJ/kg through hybrid processes that combine several up-grading variations (Ramirez et al., 2015). Based on calorific values, HTP products appear to be able to compete with conventional fuels. HTC coal even achieves higher calorific values than raw lignite. In addition to using HTP products for energetic purposes, also other fields of application for HTP products are conceivable. For example, the use of hydro-coal/bio-char as a soil conditioner with integrated carbon sequestration in the soil appears promising (Chan et al., 2007; Glowacki, 2015),

but also problems due to adverse effects on plant growth must be considered (Rilling et al., 2010). Using hydro-coal as soil amendment, as much carbon content as possible should be transferred from the feed into the hydro-coal. Generally, this varies between 70% and 75% by weight (water and ash free) which is already a considerably high value. Taking into account that a high carbon content is also an indicator of a high calorific value, these two values of hydro-coal should be maximized (Ramke et al., 2012; Vogel, 2016).

Although a high number of primary carbon is transferred to the hydro-coal, a considerable proportion is split off to the process water. The process water is therefore highly loaded with carbon and other organic compounds (especially nitrogen and phosphate) and – in particular if sewage sludge is utilized - with heavy metals, pathogens and pharmaceuticals that are split off out of the sludge (Ohlert, 2015). Table 4 shows the sum parameters for the organic contamination of HTC process water.

Table 4. TOC, COD and BOD₅ values of process water from HTC (Data from Escala et al., 2013; Ramke, 2011)

Sum parameter	Range of concentration in HTC process water
Total Organic Carbon (TOC)	9,000 – 36,000 mg C/L
Chemical Oxygen Demand (COD)	24,200 – 68,500 mg O ₂ /L
Biochemical Oxygen Demand (BOD ₅)	10,000 – 42,000 mg O ₂ /L

Solutions are currently being sought for the most efficient way to treat the process water. Discharging the process water into a wastewater treatment plant (WWTP) seems to be a simple solution. However, several batch experiments have shown that the COD values are permanently too high for the process water to be simply discharged in the wastewater regarding current legal thresholds. Most WWTP operators do not allow process water to be discharged since thresholds can be exceeded. Due to this, some studies have already investigated pretreating the process water

before discharging it. Wet oxidation and membrane processes achieved promising results for reducing pollution and thus the TOC value (up to 74%). That means that after the pretreatment of the process water a discharge into a WWTP will be possible (Loewen, 2013; Ohlert, 2015; Ramke et al., 2012; Reza et al., 2016; vom Eyser et al., 2015; Weiner et al., 2013). Another way to reduce the organic content of the process water is to separate out phosphorus. A positive side effect is that the sequestered phosphorus can be used as a fertilizer. However, such procedures have currently a low feasibility which is why they are not widespread (Remy and Stüber, 2015; Vogel, 2016).

An undesired process water also occurs during HTL although it is usually less polluted than the process water of HTC. After catalytic liquefaction (CatLiq), the TOC of the HTL process water is about 3,300 mg C/L which is considerably less critical than the TOC of the HTC process water. The gaseous phase that occurs during HTL mostly consists of carbon dioxide (~ 95%) and traces of nitrogen, hydrogen, carbon oxide and methane. Depending on the process design (e.g. hydrofaction, hydrothermal upgrading) 18% to 40% of the feed-in dry matter is split off to the gaseous phase. The carbonized organic solid material, which is another by-product of most HTL processes, is often suspended in the HTL oil. Adding alkaline salts can reduce the proportion of this solid phase (Vogel, 2016).

Undesired by-products of HTG include salts and minerals that are part of most feedstock. Most HTG designs do not separate out these materials during the process so that they occur later as an output in the process water. During processing, they often disturb the functionality of the catalyst. Some applications try to separate the salts and minerals during the process, however this is complex because of the phase reactions of the salt-water-organics mix (Müller, 2012; Schubert et al., 2010).

3.2. Economic aspects of Hydrothermal Processes

3.2.1. Feedstock supply costs

HTP feedstock supply costs consist of feedstock prices, logistic costs (collection; transport; storage) and feedstock preparation costs (drying; thickening; crushing; pressing etc.) (U.S. Department of Energy, 2014; Zhu et al., 2013). In terms of the technical biomass potential of HTP in Germany, the feedstock prices of the potential substrates - animal excreta, sewage sludge and stalk landscaping materials - are considerably low because these substrates are residues that must be disposed of in most cases, because of legal requirements like the European Waste Framework Directive (EU 2008). Studies have estimated average feedstock prices for these substrates. Leuer (2008) calculated an average feedstock price for animal excreta in Germany of between 2.17 and 2.82 EUR/ton of fresh matter. Because sewage sludge must be disposed, WWTP operators are potentially willing to pay for this. HTP is one potential disposal path. Therefore, instead of incurring costs, additional income (disposal costs for plant operators) can be generated by utilizing sewage sludge. However, this is not common practice in Germany so far, which is why a potential revenue - which usually varies between 8.00 and 12.00 EUR/t of fresh matter (Schumacher and Nebucat, 2009) - cannot be calculated (Wirth, 2017). Usually, stalk landscaping materials can be used without cost incurrence because no functioning market for such materials exists in Germany (Menzel, 2015). It should be noted that these numbers are relatively old and further investigations are recommended to generate current data on these prices. Pretreatment and conditioning only seem to be necessary for stalk landscaping material. It is essential to shred the material into small enough particles so that the substrate can be effectively pumped into the plant. Assuming that the preparation costs for landscaping materials used in HTP are similar to those used for biogas production, they range between 4.50 to 5.60 EUR/ton of fresh matter depending on costs for personnel (Leible et al., 2015).

3.2.2. Investment and operating costs

Investment costs (building; equipment; site development) and operating costs (operating material costs; staff costs; maintenance costs; insurance costs) highly depend on the individual business case. Influencing factors can be plant location, composition of the substrates used, scale of the plant, energy and mass balance of the process (especially the proportion of process water related to the product output), process design and calibration of process parameters (AVA CO2 Schweiz AG, 2012; Eberhardt et al., 2011; U.S. Department of Energy, 2014). The process energy balance significantly influences the energy costs - an important part of the operating costs (Buttmann, 2011; Kruse, 2008). Three important aspects must be considered:

(1) The amount of heated water: To reduce the energy that is necessary to heat the water, using substrates that have a dry matter content of around 20-30% is recommended (Greve et al., 2014).

(2) The loss of energy through dissolved by-products: To reduce this loss, a maximum amount of the process water must be recovered and later used for energetic purposes (for example, when process water was used in biogas plants there was a 19% gain in energy efficiency compared to the reference state) (Greve et al., 2014).

(3) The exothermic process conditions: To increase the overall energy efficiency, a maximum amount of waste heat must be used during the process. Current studies have shown that up to 90% of the required process heat can be supplied through waste heat (Greve et al., 2014; Kruse, 2008; Remy and Stüber, 2008).

Estimating the investment and operating costs for large-scale HTP plants in Germany is difficult because there is a lack of experience with such installations (Vogel, 2016) even that some scenarios for large-scale HTC plant concepts already exist (Child, 2014). Hence, some calculations for these

cost components are available. Figure 4 shows the overall investment costs and annual operating cost calculation for sample case studies compared to conventional reference systems.

Table 5. Case studies representing sample investment and operating costs of HTP plants

Case study / reference case	Substrates	Plant scale	Reference
Case 1: AVA-CO2 commercial HTC plant	Sewage sludge with 25% dry matter content	80,000 t/a	AVA CO2 Schweiz AG (2014)
Case 2: TerraNova energy pilot HTC plant	Sewage sludge with 20% dry matter content	5,000 t/a	TerraNova energy GmbH (2011)
Case 3: Modeled HTL and integrated catalytic hydrothermal gasification (CHG) plant	Sewage sludge with 12% dry matter content	36,500 t/a	U.S. Department of Energy (2016)
Reference 1: Commercial sewage sludge mono-incineration plant	Sewage sludge with 25% dry matter content	30,000 t/a	Glatzner and Friedrich (2015)
Reference 2: Commercial biogas plant	Corn silage with 32% dry matter content	9,000 t/a	TerraNova energy GmbH (2011)

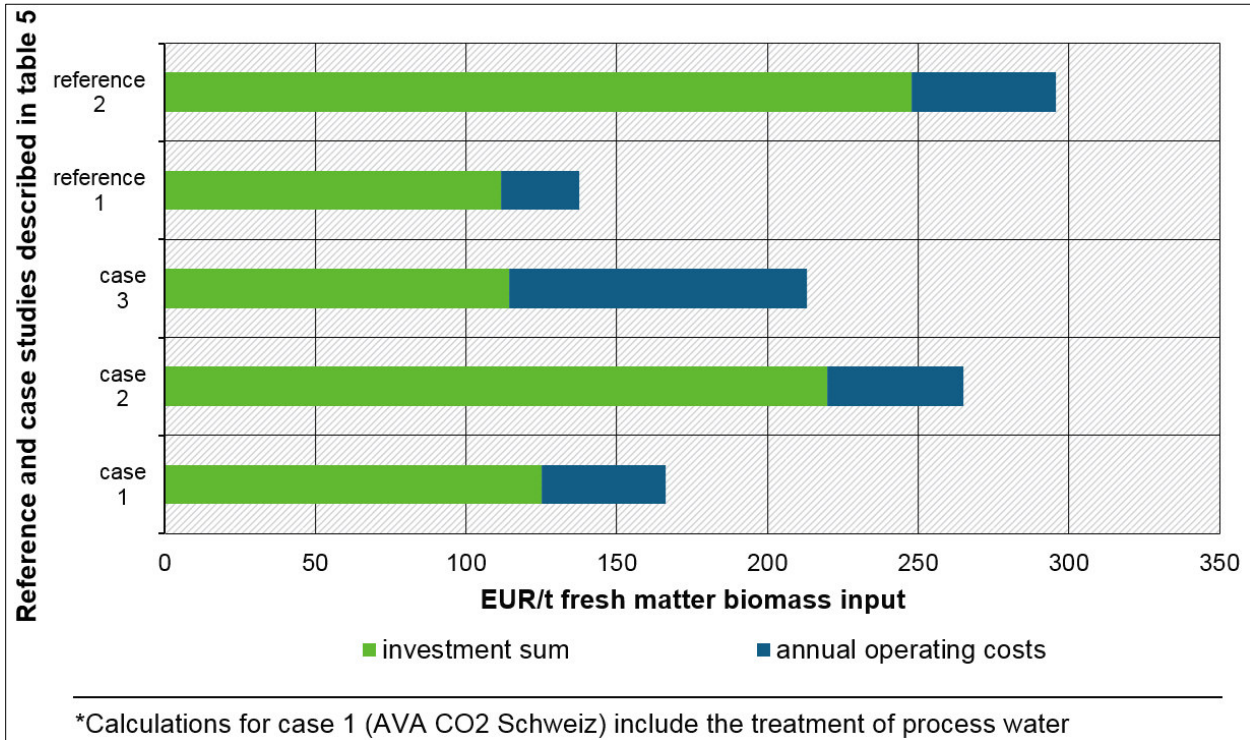


Figure 4. Investment costs (no annuity) and annual operating costs in EUR per ton of fresh matter biomass input for HTP sample plants compared to conventional reference plants (Data from AVA

CO₂ Schweiz AG, 2014; Glatzner and Friedrich, 2015; TerraNova energy GmbH, 2011; U.S. Department of Energy, 2016)

In terms of investment costs, large-scale plants have lower costs per ton of dry matter biomass input, which is attributed to economies of scale (Carlino, 1978). No such connection can be drawn for operating costs because they are comparable in both the smallest plant (5,000 t/a) and the largest plant (80,000 t/a). Figure 4 shows that the investment and operating costs of different HTP cases (explained in Table 5) appear to be able to compete with conventional technologies.

3.2.3. Transport and distribution costs

Distribution costs occur when the HTP products are moved to resellers and customers. Cost components may include warehousing costs, transport and logistic costs, or reclamation costs (Springer-Gabler, 2017). The costs are highly dependent on the individual business case and difficult to assess in general. The transport costs have a significant influence on the overall process chain economy. In general, the distance between the location of where the substrate occurs, HTP plant, and the location of the customer is proportional to the increase in transport cost (Eberhardt et al., 2011). The main cost components are staff costs (40 - 50%), capital costs, energy/fuel costs and costs for maintenance and insurance (10 - 20% respectively) (Gasafi et al., 2008). Costs for transporting the HTP products (HTL coal, bio-char, HTL oil, HTG gas) to the customer are directly linked to the transport vehicle used (e.g. fuel consumption), the transport distance, the time for loading and unloading, the density of the product (kg/m³), and the volume of the transport container (m³) (Eberhardt et al., 2011).

3.2.4. Sales

The sales markets for HTP products are diverse and include energy production, fertilizing and soil conditioning, chemical production, and material applications. Based on the focus of most research projects (Ardissone, et al. 2015; Berge, 2015; TerraNova energy GmbH, 2011; Zhengang, 2012) and the main products of current installations in practice for all HTP products, the market for energy production seems to be the most promising. The market for soil conditioners is also highly significant for the HTC product bio-char (Glowacki, 2015). Experts estimate also a high potential of HTC products for material applications in future (Titrici et al., 2012; Wirth, 2017).

In energy production, HTP products can be sold to power plants (e.g. fuels) and to industry (e.g. co-incineration). The income from the sale of these products is enhanced by the additional savings that arise from the fact that no emission allowances are necessary for these fuels. However, it must be examined whether energetic HTP products can be utilized within the existing plant and industry infrastructure, or whether reconstruction measures are necessary (Eberhardt et al., 2011). The most important factor for most potential HTP fuel product users is the price (production costs plus profit margin) of the HTP fuel compared to conventional fossil or biogenic fuels. Figure 5 shows some examples of the production costs of different HTP fuels based on specific plant concepts compared to average fossil fuel production costs (data from Zeymer et al. (2015)) including additional prices for emission allowances and biogenic synthetic natural gas (bio SNG data from Billig (2016)). The production costs are based on all previously described cost components whereby the investment costs are calculated using the equivalent annual cost method (cf. Edge and Irvine, 1981). Table 6 describes the HTP plant concepts.

Table 6. Characteristics of HTP plant concepts as examples for HTP production costs

Plant concept	HTC 1 (present)	HTC 2 (present)	HTL (modeled)	HTG (modeled)
Substrates	Municipal bio-waste	Fermentation residues	Chlorella algae	Corn stover
Plant scale	2,500 t/a	2,500 t/a	489,100 t/a	858,480 t/a

Additional product treatment steps	Pelletizing and packaging	Free heat delivery	Including HTL-oil upgrading step	Pretreatment and hydrolysate conditioning of substrate
Product	HTC coal dust	HTC coal dust	HTL oil of fuel quality	Synthetic Natural Gas (SNG)
References	Hallesche Wasser und Stadtwirtschaft (2015)	Hallesche Wasser und Stadtwirtschaft (2015)	U.S. Department of Energy (2014)	U.S. Department of Energy (2009)

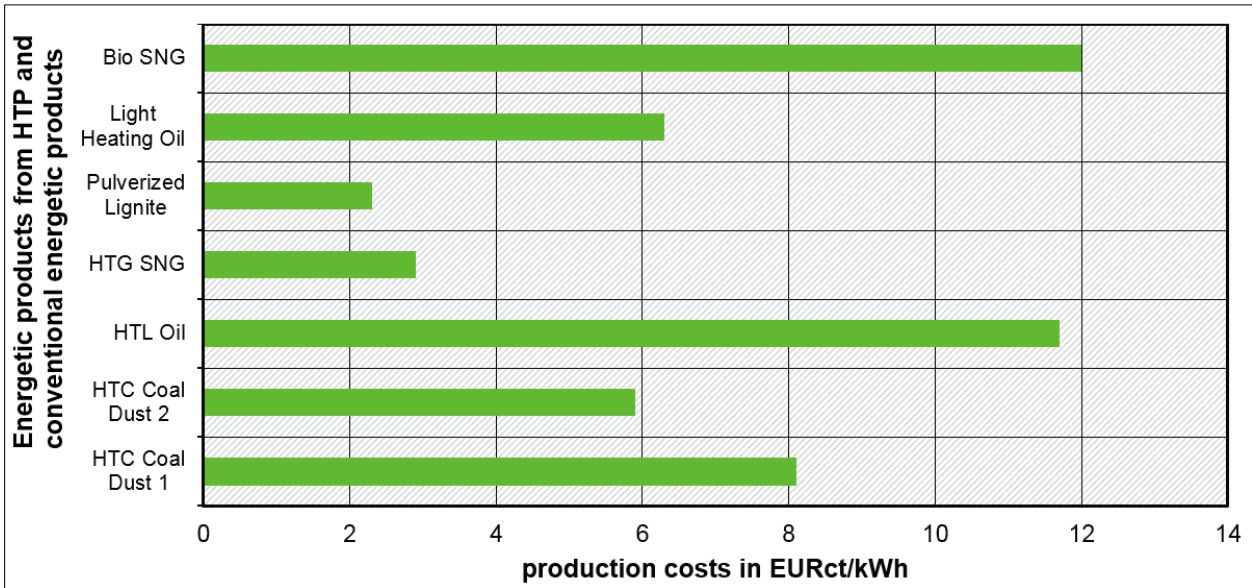


Figure 5. Production costs in EURct/kWh of sample HTP energetic products compared to conventional fossil and biogenic energy products (Data from Hallesche Wasser und Stadtwirtschaft, 2015; U.S. Department of Energy, 2009; U.S. Department of Energy, 2014)

Currently most energetic HTP products are more expensive in production costs than conventional fossil alternatives. This is due to the novelty of the technology platform and lack of experience with large-scale applications (e.g. absence of learning curve effects). However, it has been noted that production can compete with Bio-SNG. The markets for soil conditioners are also highly relevant to HTC because bio-char can be used for this purpose. The production costs for

HTC char lie between 75 and 100 EUR per ton, which is comparable to conventional soil conditioners like peat (Top Agrar Online, 2011).

3.3. Environmental issues of Hydrothermal Processes

The Life Cycle Assessment (LCA) is the most common method used to analyze environmental effects including greenhouse gas emissions (GHG), toxicity, or eutrophication along the entire process chain (see the illustration of the process chain in Fig. 2). Also several LCA have been carried out with respect to HTP (e.g. Ahamed et al., 2016; Benavente et al., 2017). To illustrate this, Figure 6 shows LCA results for greenhouse gas emissions (GHG) of specific HTP concepts compared to conventional reference systems. Table 7 briefly describes the specific concepts, i.e. HTC using green-waste, HTL using microalgae and HTG using manure as substrate.

Table 7. Characteristics of HTP plant concepts as examples for HTP greenhouse gas emissions

HTP type	Substrate	System scope	Reference system	References
HTC	Green-waste	Green waste from green fields → feedstock supply → HTC processing → pelletizing of HTC coal → transportation and incineration in a 30 kW pellet stove → <i>heat</i>	Heat generation through natural gas boiler	Hallesche Wasser und Stadtwirtschaft (2015)
HTL	Microalgae	Algae growth → dewatering → HTL processing → bio-oil stabilization → conversion to renewable diesel (upgrading) → transport and distribution → <i>mobility</i>	Conventional diesel	Bennion et al. (2015)
HTG	Manure	Manure preparation and conditioning (ultrafiltration) → HTG processing → production of SNG → <i>heat and electricity</i>	Anaerobic digestion	Luterbacher et al. (2009)

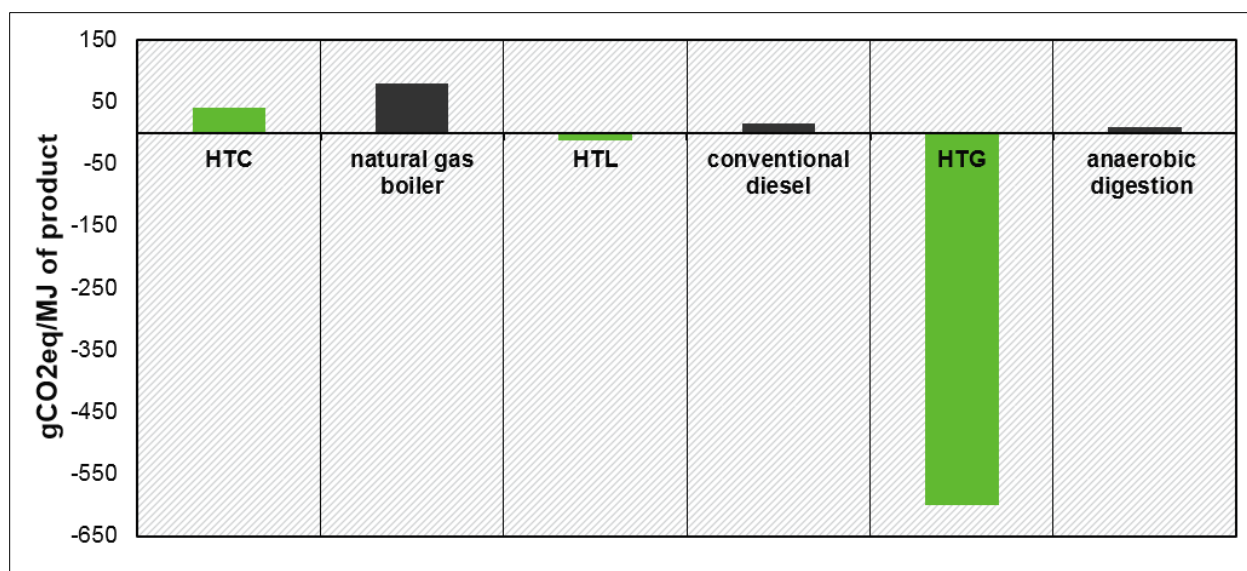


Figure 6. Carbon dioxide equivalent per mega joule of sample HTP concepts compared to conventional reference systems (Data from Bennion et al., 2015; Hallesche Wasser und Stadtwirtschaft, 2015; Luterbacher et al., 2009)

The GHG balances of the sample HTL and HTG concepts seem especially promising because they are in fact even negative. In the case of HTL, carbon dioxide binds to algae during the growth phase generating additional GHG credits. In addition, the process water from HTL contains ammonium and phosphate which can be used as a growing medium for algae. This makes mineral growing media superfluous and cuts out additional emissions. Furthermore, the gaseous by-product of HTL (especially hydrogen and methane) is simultaneously combusted to improve the energetics of the system, which saves even more greenhouse gases (Bennion et al., 2015). However, other studies have shown that the installation and use of infrastructure equipment, like HTL upgrading technology, creates a significant energy burden which is very relevant for the overall environmental impact (Ramirez et al., 2015; U.S. Department of Energy, 2014).

In the case of HTG, the high potential for GHG savings is mainly the result of the use of manure, which is a problematic biomass when it comes to greenhouse gas emissions. Manure emits nitrous oxide, which has a global warming potential that is 310 times higher than that of carbon dioxide. Hence, a considerable amount of GHG is saved through the treatment of this biomass as part of the HTG process (Luterbacher et al., 2009).

HTC has a high potential for additional carbon credits by binding carbon to soil using bio-char as a soil conditioner. When indirect effects are also taken into account (e.g. decreased GHG emissions due to a lower production of mineral fertilizers), the overall carbon mitigation potential increases further. According to recent research, the most influential factors for the potential of bio-char to mitigate carbon include the amount of carbon applied with char, additional soil organic carbon, and indirect carbon credits (e.g. the need for fewer mineral fertilizers which is also relevant for other HTP products due to nutrient recovery from process water) (Libra et al., 2011; Luterbacher et al., 2009). However, the long-term stability of hydro-coal in soil has yet to be sufficiently investigated.

When bio-char is added to the soil, CO₂, N₂O and CH₄ soil emissions must be taken into consideration. N₂O emissions are reduced after the application of the char (Lehmann, 2007; Singh et al., 2010; van Zwieten et al., 2010). CO₂ soil emissions are also lower (Lehmann, 2007; Lu et al., 2012; Singh et al., 2010; van Zwieten et al., 2010). In contrast, higher emissions of methane were recorded after bio-char was added to soil (van Zwieten et al., 2009).

In addition to GHG emissions, environmental impact issues, like acidification, human-toxicity, eco-toxicity, and resource depletion or eutrophication, are important if there is to be a holistic assessment of the environmental burden connected with the processes, products and services (Berge et al., 2015; Krebs et al., 2015; Lu et al., 2012). For example, a study of Berge et al. (2015)

focusing on food waste HTC (275 °C, 16 h and 32% dry matter content) concluded that hydro-coal combustion has the most beneficial influence on the environmental impact (GWP: -99%, acidification: -93% and human-toxicity, non-cancer: -38%) when compared with the use of conventional lignite. In contrast, the process water emissions have the most adverse environmental impact (> 60% for human-toxicity, eco-toxicity and freshwater eutrophication). Krebs et al. (2015) evaluated the overall environmental burden of sewage sludge HTC on an industrial scale and show that the process is environmentally promising under specific conditions. These include when waste heat or other local renewables are used in the processing, phosphorus and nitrogen content is reduced in the process water, phosphorus is recovered, HTC coal is used as a substitute for fossil fuels and HTC replaces sewage sludge drying that uses conventional fuels. Most studies generally conclude that HTP has a considerably lower Global Warming Potential (GWP) than comparable conventional fossil and biogenic processes (Berge et al., 2015; Clarens et al., 2013; Krebs et al., 2015; Lehmann, 2007; Ramirez et al., 2015; U.S. Department of Energy, 2014).

3.4. Legal issues surrounding Hydrothermal Processes in Germany

3.4.1. Legal regulations affecting feedstock supply

When examining the suitable HTP feedstock available in Germany (animal excreta, sewage sludge and stalk landscaping materials), the utilization of sewage sludge, in particular, is subject to strict regulations. An amendment to the German Sewage Sludge Ordinance (Klärschlammverordnung (AbfKlärV)) means that the thresholds for agricultural utilization of sewage sludge were tightened, resulting in the loss of a central line of business for many sewage sludge disposal companies. According to this amendment, only sewage sludge from WWTP with a maximum of 50,000 inhabitant-equivalents may be used for agricultural purposes (BMUB,

2016a). In addition, the thresholds of the German Fertilizer Ordinance (Düngemittelverordnung (DüMV)) already impede agricultural usage for some forms of sludge (DüMV, 2012; Greve et al., 2014; Libra et al., 2011). Hence, Germany is currently urgently in need of new, sustainable ways of treating sewage sludge. For instance, co-incineration is not promising for the utilization of sewage sludge because of unavailable capacities of appropriate technical facilities for this purpose in Germany and the release of phosphorous during the incineration process (Glowacki, 2015). Though the new sewage sludge ordinance regulates phosphorous recycling of sewage sludge that exceeds certain phosphorous thresholds, the co-incineration of sludge with high P-values is nevertheless permitted (Greve et al., 2014). Therefore, only a few sewage sludge treatment options remain. Incineration is a suitable energetic treatment option but the energy efficiency is low as a result of an energy-intensive pretreatment process (thickening, drying) of the sludge. With this in mind, HTP seems promising from a legal perspective.

The use of animal excreta through biomass conversion processes has been promoted for several years through regulations like the German Renewable Energy Act (EEG, 2017). Even though the funding schemes of this regulation has been on applied to anaerobic processing, these efforts have shown there is a legal intention to sustainably process animal excreta.

Stalk landscaping materials are defined as bio-waste according to the German Law on Closed Cycle Management and Waste (KrWG § 3 sec. 7 no. 2). This legal scope can be disregarded if the stalk landscaping material consists mostly of logs and huge knots that are used for energetic purposes. However, landscaping material that is suitable for HTP does not fulfil these requirements (Kehres, 2012).

3.4.2. Process and plant standards

Most HTP applications currently operate as pilot or demonstration plants (cf. Boukis et al., 2005; Boukis et al., 2008; Remy et al., 2013). Furthermore, there is a wide range of potential process designs and the optimal calibration of process parameters and other important influencing factors are currently not fully known. This explains why process or plant standards for HTP do not exist so far (Greve et al., 2014). However, they need to be developed in order to reduce uncertainties for technology investors and policy makers (e.g. for funding decision and legal regulations) as well as to enhance the acceptance for the technologies in society.

3.4.3. Product quality standards and product authorization

Standardizing the product quality is highly relevant to increase legal certainty for HTP stakeholders, especially product user, because HTP products become comparable to each other and to other similar products through this. Hence, quality standards governing feedstock, production conditions, product composition and physical, chemical and biological characteristics are already in discussion (Libra et al., 2011). Efforts are already underway to establish quality standards for the use of bio-char as a soil conditioner (e.g. from HTC). The guidelines on the production of bio-char (European Biochar Certificate), which were developed in 2012, define them as materials used for sustainable agriculture produced through pyrolysis with an oxygen content of less than 2% and at temperatures of between 350 and 1000 °C (EBC, 2012). Any bio-char that is not a product or co-product of pyrolysis is regulated as waste in accordance with the European Waste Framework Directive (2008/98/EC). In addition to the European regulations, potential bio-char applications must also be in accordance with national legislation that often defines threshold limits for bio-char based on specific substrates (e.g. sewage sludge). However, the EBC is an initiative and has yet to be officially implemented by European legislation (only Switzerland has officially implemented the EBC). The lack of legislation needs to be clarified before a bio-char market can be implemented

(Montanarella, 2013). In Germany, the German Fertilizer Ordinance (Article 4 Appendix 2 in connection with Table 7 DüMV) regulates the use of bio-char as a soil conditioner. HTP products are not listed as products according to DüMV, complicating their admittance as soil conditioners. Because no robust data and information regarding the long-term stability of hydro-coal in soils is available, it seems very unlikely that hydro-coal will be allowed to be used as a regular fertilizer in line with DüMV anytime soon (Greve et al., 2014).

Fuels based on sewage sludge are defined as waste in Germany because of the high level of contamination of the raw material. Thus, they can only be used in waste incineration plants or waste co-incineration plants in accordance with the 17th Federal Emissions Control Act (BImSchV). This is a legal issue because fuels from HTP are also not defined as products according Section 3(1) of the first BImSchV and are legally regarded as waste. Hence, energy-intensive companies have no demand for such fuels because they cannot use them in conventional plants as substitute fuel (Gawel et al., 2015). However, based on Section 5(1) of the German Law on Closed Cycle Management and Waste (KrWG), bio-based fuels from substrates that are not contaminated with pollutants can be used as fuels. Therefore, the legal barriers are highly relevant, especially for fuels based on sewage sludge. European legislation initiatives have already tried to change the legal basis for fuels based on sewage sludge. Recommendations for changing the EU Waste Framework Directive have been put forward including the suggestion of allowing sludge-based fuels that have undergone treatment in a refinement process (2008/98/EC).

4. Discussion

4.1. Potentials and barriers for HTP in Germany

511 Based on the information of the previous sections, the following key potentials and barriers were
 512 identified as shown in Table 8.

513 **Table 8.** Potentials and barriers for HTP in Germany

Potentials	Barriers
<i>Technology</i>	
<ul style="list-style-type: none"> • Mobilization of unused wet and sludgy biogenic residues • Faster processing than other biogenic treatment options • Process efficiency higher than conventional biogenic treatment options (Table 3) • Calorific values of energetic HTP products are competitive with conventional fuels (Figure 3) • High carbon content in hydro-char • Parallel phosphorus recycling from process water • Combination of HTP and wastewater treatment plants (e.g. use of sewage sludge and recovery of process water) 	<ul style="list-style-type: none"> • Lack of experience with large-scale commercial applications (e.g. learning curve effect) • Lack of knowledge regarding: <ul style="list-style-type: none"> ○ Process kinetics ○ Optimal calibration of process parameters • No optimal solution for the treatment of the highly contaminated process water of HTC
<i>Economy</i>	
<ul style="list-style-type: none"> • Low feedstock supply costs (Table 5) due to: <ul style="list-style-type: none"> ○ Low overall substrate costs ○ Potential on additional revenues through the use of sewage sludge (disposal costs) ○ Low substrate preparation costs • Competitive investment and operating costs are expected which compare with conventional biogenic treatment options (Figure 4) 	<ul style="list-style-type: none"> • Lack of cost data for large-scale commercial plants (due to a lack of experience) • Higher productions costs are expected for HTP fuels than for conventional fuels (Figure 5)

<ul style="list-style-type: none"> • Production costs of HTC-char expected to compete with conventional soil conditioners 	
<i>Environment</i>	
<ul style="list-style-type: none"> • Significantly lower GWP of HTP possible compared to conventional reference systems • HTC-char as carbon sink in soil 	<ul style="list-style-type: none"> • Little knowledge about stability of HTC-char as a carbon sink in soil • Negative environmental burden of contaminated HTP process water
<i>Legislation</i>	
<ul style="list-style-type: none"> • Strict legislation for the utilization of sewage sludge for agriculture enhances need for alternative treatment paths like HTP 	<ul style="list-style-type: none"> • HTP products not authorized as fuel or fertilizers (waste characteristic) • A lack of standards and norms for HTP products and the processing itself increases uncertainties for stakeholders

514

515 4.2. Future research needs

516 Future research is necessary to solve fundamental problems (highlighted in red in Figure 7) and
517 to foster the most important potentials (highlighted in green in Figure 7). Figure 7 provides an
518 overview of all relevant research areas (according to Figure 2) and connects them to the current
519 state of knowledge based on the information in the review. Fundamental research is necessary for
520 the research areas categorized in red whereby application-oriented research is recommended for
521 the areas marked green. Further research is recommended for the areas marked amber but they
522 have a lower priority than the other two areas.

Research Topic		Current State of Knowledge
Technology		
Suitable substrates and biomass potential		mostly known
Necessary pre-treatment of substrates		partly known
Process parameter calibration and process design		knowledge gaps (esp. large scale)
Resulting products and product usage		mostly known
Treatment of by-products		knowledge gaps
Economy		
Feedstock supply costs		mostly known
Investment costs		knowledge gaps (esp. large scale)
Operating costs		knowledge gaps (esp. large scale)
Distribution costs		knowledge gaps
Sales of HTP products		partly known
Environment		
Life Cycle Performance		partly known
Legislation		
Legal status of feedstock supply		partly known
Process and plant standards		missing
Product standards		mostly missing
Clear regulations for product authorization		missing

	Primary application-oriented research necessary
	Partly application-oriented and fundamental research necessary
	Primary fundamental research necessary

Figure 7. Future HTP research needs

Especially, the research gaps that are highlighted in red need a special attention because fundamental research is still necessary for them. To fulfil these gaps, knowledge building is most important. For example, for the treatment of by-products like polluted process water several solution exist as shown in section 3. However, to identify the most optimal solution or combination of solutions it will be necessary to develop theoretically based decision making tools as well as to enable a practical in-field application to verify if the theoretically selected solutions are also operational. Such processes need the involvement of several stakeholders (e.g. technology developers, technology user, product users, retailers, policy makers, researchers) that must share experiences and knowledge.

5. Conclusion

Hydrothermal Processes are an appropriate technology platform for mobilizing currently unused biogenic waste residues in Germany, however several technological, economic, environmental and legal questions have to be considered as Table 8 and Figure 7 show. HTP are promising regarding their ability to mobilize wet and sludgy biogenic residues that are currently unused and partly subject to disposal pressure (e.g. sewage sludge). Furthermore, HTP products are able to compete with conventional reference products in terms of their calorific value (energy production) and carbon content (fertilizing). Their advantages include being notably more efficient than other technologies that use wet substrates, having the potential to provide an additional biotechnology for existing treatment facilities, and having a lower climate footprint than comparable technologies. However, barriers still exist which could impede the successful implementation of HTP in Germany. Fundamental and applied-oriented research is needed to achieve the next level of technological readiness. This includes building upon knowledge of process kinetics and process design, the treatment of by-products, and the cost structure of the entire process chain. Insufficient knowledge about HTC process water treatment leads to high costs (e.g. use of expensive and inefficient treatment options), environmental problems (e.g. process water emissions, contaminant influx) and legal restrictions (e.g. thresholds for wastewater discharge to WWTP). Furthermore, the lack of data on large-scale investments increases the uncertainties for many potentially interested investors, whereby this problem is a result of a general absence of large-scale applications. The categories under scrutiny are intertwined with one another. For example, the lack of knowledge on the long-term stability of hydro-char in soils - mainly an environmental problem - reduces its economic chances due to rising uncertainties on the markets for soil conditioners.

The current legal situation in Germany is both a blessing and a curse. Through the amendment of the German Sewage Sludge Ordinance, new treatment options for sewage sludge are urgently needed. Because sewage sludge is one of the most suitable inputs for HTP this is a general advantage. At the same time, restrictions by BImSchV and DüMV lead to a situation where HTP products from sewage sludge are generally not permitted as regular fuel and soil conditioners.

More research on HTP is necessary to reduce the technological, economic and environmental barriers. A better holistic understanding of this technology platform will help to generate a basis of argument for legal adjustments, will be necessary to enable a successful large-scale application of HTP in biogenic waste management in Germany. Regarding the potential of HTP to utilize sewage sludge, it appears that the current legislation can be adapted in order to simplify the application of HTP for the treatment of sewage sludge. With regard to the current amendment of the Sewage Sludge Ordinance, this could help to put sludge onto an efficient and value-enhancing treatment pathway that meets the new legal requirements.

Acknowledgements

We are grateful to Romann Glowacki (DBFZ) for all the fruitful discussions about current research activities regarding HTP, and for his intermediary role to reach actors within the “BMBF Innovationsforum Hydrothermale Prozesse”. We thank the anonymous reviewers for their critical analyses and comments that helped in finalizing our manuscript.

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