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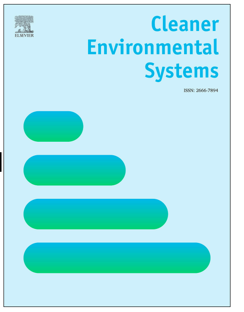

Life cycle assessment of biochar and hydrochar derived from sewage sludge: Material or energy utilization?

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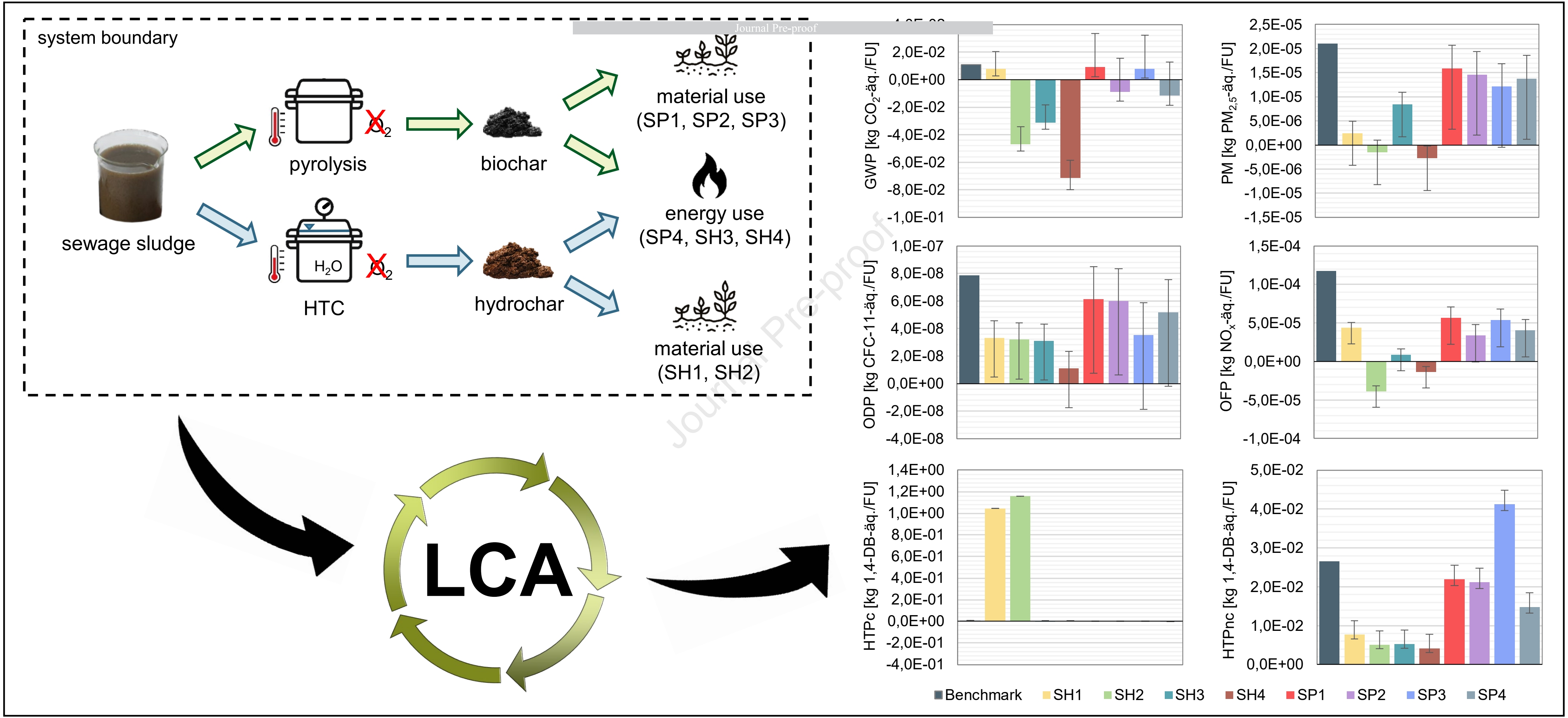

system boundary
sewage sludge
pyrolysis
O_2
biochar
HTC
H_2O
O_2
hydrochar
material use
(SP1, SP2, SP3)
energy use
(SP4, SH3, SH4)
material use
(SH1, SH2)
LCA
GWP [kg CO_2-äq./FU]
PM [kg $PM_{2,5}$-äq./FU]
ODP [kg CFC-11-äq./FU]
OFP [kg NO_x-äq./FU]
HTPc [kg 1,4-DB-äq./FU]
HTPnc [kg 1,4-DB-äq./FU]
Benchmark
SH1
SH2
SH3
SH4
SP1
SP2
SP3
SP4

Abstract: In this work, alternative process chains for sewage sludge treatment by hydrothermal carbonization (HTC) and pyrolysis and the subsequent utilization of biochar and hydrochar were investigated by means of a comparative life cycle assessment (LCA). The mono-incineration of sewage sludge was defined as the benchmark process, being the main valorization route in several countries. The results revealed an advantage for HTC compared to pyrolysis and mono-incineration in most environmental impact categories, thanks to the higher char yield and the lower energy request. The global warming potential (GWP) of using hydrochar ranged from −71.4 to 7.7 g CO_2-eq. kg^{-1} sewage sludge. However, the direct material application of the hydrochar in agriculture showed an increased toxicity potential, so that an energy utilization appears to be more environmentally friendly. For the pyrolysis route, a slightly higher energy demand and thus a higher environmental impact was determined, whereby GWP was between −11.7 to 9.1 g CO_2-eq. kg^{-1} sewage sludge. The direct material application of the biochar in agriculture also showed low toxicity potentials while achieving nutrient recycling for phosphorus and a long-term carbon sequestration potential. Overall, ecological advantages were demonstrated for both thermochemical processes, making pyrolysis and HTC promising alternatives for sustainable sludge management.

Keywords:
sewage sludge utilization, nutrient recycling, environmental impact, carbon sequestration, pyrolysis, hydrothermal carbonization

Word count: 7754

1 Introduction

In recent decades, the increase in the world's population and global economic growth has led to a higher demand for resources, drastically raising the volume of produced wastewater and the resulting environmental pollution [1]. For this reason, wastewater treatment plants are built and operated to purify wastewater and minimize damages to the environment and human health [2]. The main by-product of each wastewater treatment process is sewage sludge, which contains valuable nutrients such as nitrogen and phosphorus that make it suitable for use as a fertilizer [3–5]. The agricultural reuse of sewage sludge could theoretically replace up to 50% of the mineral phosphate fertilizer that is applied annually in European agriculture [6]. A long-term strategy is therefore required to enable waste-based nutrient recycling, especially for critical raw materials such as phosphorus.

At the same time, however, sewage sludge also contains potentially harmful substances such as heavy metals, organic pollutants, and pathogens that can accumulate in the receiving soils [7], requiring the development of alternative treatment and recycling processes [8–10]. One of these possibilities is sewage sludge incineration (i.e., combustion with energy recovery). However, the high water and ash content of the sludge leads to an overall low calorific value, requiring energy-intensive pre-treatment to enable autothermal combustion [11]. Nevertheless, new facilities for sewage sludge mono-incineration are currently being built, despite significant technical and economic uncertainties regarding phosphorus recovery from ashes, as the necessary processes have not yet been widely established on an industrial scale [12–15].

Given the current growing demands for reducing pollutant and greenhouse gas (GHG) emissions, recovering nutrients, generating clean energy and implementing circular economy principles, it is important to rethink the conventional ways of utilizing sewage sludge to tap into its largely unexploited environmental and economic potential [1]. As an alternative to mono-incineration, different thermochemical processes, in particular pyrolysis and hydrothermal carbonization (HTC), have recently been tested at the laboratory or pilot scale, and even in some full-scale facilities [5,14,16]. Pyrolysis consists of an endothermic conversion of complex substances in an oxygen-free environment, and is normally performed at 500-700 °C, while HTC takes place in aqueous phase and at lower temperatures (150-350 °C), and thus is specifically adapt to wet biomasses, as it does not require an intense preliminary drying step [4,17]. Through thermochemical processes, the sludge is mostly converted to solid products called biochar or hydrochar, which can be used as agricultural amendments, fuels, additives to the anaerobic digestion (AD), or even adsorbents for gaseous pollutants [17].

However, the ecological effects of these alternative treatment processes (HTC and pyrolysis) are still largely unexplored. It is unclear whether the alternative process chains offer ecological advantages over mono-incineration and which application paths for the resulting biochar and hydrochar will enable a sustainable circular economy for critical resources while achieving the lowest possible emissions profile [11,18]. Recently, it was demonstrated that biochar and hydrochar produced from straw and manure positively influence soil physicochemical properties with hydrochar outperforming biochar thanks to GHG emission reduction and degradation of cellulose, lignin and chitin [19]. However, the results strongly depend on the specific characteristics of the processed biomass.

Therefore, a thorough environmental assessment is urgently required to identify the most suitable treatment method for sewage sludge, including the subsequent use of the resulting products, providing useful indications for stakeholders. In this sense, life cycle assessment

(LCA) can help to broaden the perspective beyond waste management. The environmental impacts of waste management often depend more on the effects on surrounding systems than on the emissions from waste management itself [20]. Consequently, the focus on the development of resource and waste management strategies has recently shifted from weight-based recycling targets to impact-oriented measures [21]. LCA is now ubiquitous as an integrative component for further development of waste management and a virtuous utilization of natural resources, including the adjacent energy industries [22–26]. Thanks to circular economy principles, the need to produce useful products or energy from sewage sludge through thermochemical conversion has become more attractive and has been investigated in several LCA studies. For example, Huang et al. (2022) [27] investigated different options to produce activated carbon (AC), biochar or bio-oil from sewage sludge by fast pyrolysis, while Xia et al. (2024) [28] examined various feedstock-based biochars as soil amendments with an LCA approach. Mannarino et al. (2022) [29] evaluated the environmental performance of sewage sludge HTC, and Ogunleye et al. (2024) [30] reviewed different hydrochar utilization pathways through their life cycle performance. However, most of the studies focus exclusively on pyrolysis [27,31] or HTC [9,29] of sewage sludge, while a fruitful comparison of HTC and pyrolysis, with useful insight to stakeholders, is seldom performed [11], requiring further studies on this outstanding topic.

In fact, to the authors' knowledge, there are no studies to date that evaluate and compare the environmental impact of different thermochemical treatments of sewage sludge, including the direct material or energy utilization of the resulting materials beyond landfilling. In this article, a LCA is therefore carried out in which the environmental impacts of various thermochemical sewage sludge treatment processes are evaluated by considering eight different recycling routes of the resulting biochar (agriculture, horticulture, AD amendment + agriculture, co-incineration) and hydrochar (agriculture, horticulture, mono-incineration, co-incineration), comparing the results with the conventional mono-incineration process. Finally, a scenario and a sensitivity analysis are performed to determine the influence of energy supply, process water treatment and flocculant type on the model results and to understand the sensitivity of the LCA results to key input parameters. The results of this study can be used to integrate environmental aspects into the early development stages of a sustainable framework for sewage sludge treatment with valuable resource recovery, as it allows optimal process selection and thus contributes to sustainable decisions in waste management strategies.

2 Materials and methods

2.1 Modelling Framework

The LCA was conducted in accordance with the ISO 14040:2006 standard [32], using the software GaBi from Sphera Solutions Inc. (Version 10.6) to model and compare the process chains. In the context of LCA application to new technologies, the main objective is typically to apply a consistent approach that includes substituted systems and identifies marginal processes directly related to the modeled processes [33]. In the ISO 14044:2006 standard [34], this method is referred to as a system extension, as the further fate of the by-products and the resulting changes (substitutions) are not cut off by the allocation [35,36].

2.1.1 Goal and Scope

The definition of the assessment goal serves as a guideline for all aspects of the scope of the study, providing the framework for the life cycle inventory (LCI) and the resulting life cycle impact assessment (LCIA) [37]. Given that an LCA is particularly useful to investigate new technologies or systems for which only limited or no data is available at an industrial scale [38], this paper investigates some relatively novel thermochemical conversion technologies. The LCA results can be used to determine whether the technologies under consideration represent an ecological improvement compared to existing technologies or whether the latter already represent the best solution. The focus is twofold: choosing the optimum disposal strategy in terms of energy and materials, and closing material cycles while minimizing pollutant emissions into the environment. The study compares pyrolysis and HTC with mono-incineration, but other new or hybrid technologies for sludge treatment, which were not the subject of this study, could also offer competitive advantages. Moreover, although many environmental impact categories were considered, the LCA might not fully encompass all potential environmental impacts, such as social and economic externalities or impacts on biodiversity, which are equally important in assessing overall sustainability.

2.1.2 Functional Unit

The LCA of a waste treatment system usually focuses on the end-of-life phase of a product or waste, considering only the processes required to manage this waste [25,39]. The functional unit is therefore defined based on the system input, in this case, the amount of sewage sludge to be disposed of [39]. The functional unit (FU) was thus defined as "treatment of 1 kg of sewage sludge after anaerobic digestion with a total solids (TS) content of 5% and a volatile solids (VS) content of 48% TS" for all systems under investigation. The

TS and VS content were derived from the mono-incineration process used by the Ecoinvent database, which served as the benchmark treatment.

2.1.3 Definition of the system boundaries

The waste treatment system evaluated in this work included different thermochemical treatment processes for digested sewage sludge as a waste product. Therefore, the previous life cycle stages of the wastewater treatment can be omitted from the assessment [35], and the sewage sludge enters the modeled system after the anaerobic digestion stage without emissions from the upstream processes [20,39]. For all other material and energy flows associated with the investigated process chains, the complete cradle-to-grave life cycles were modeled. When modeling the individual process chains based on the assumptions made and literature results, an effort was made to comprehensively record the relevant material and energy flows, ensuring that at least 95% of these flows were accounted for. To keep the balancing effort within a manageable framework, operations that were not used exclusively for the process chains under consideration were cut off. Examples include infrastructure and investment items, such as the life cycle of the roads used to transport the chars or the manufacture of the tractor used to spread the char. Many studies have shown that the emissions from reactor manufacturing and plant components over the entire life cycle are low compared to the emissions from energy provision, and therefore show limited environmental impact [2,22,25,40]. Therefore, the materials for the reactors and system components were not part of this study.

The modeled system includes sludge treatment by HTC and pyrolysis as well as four different use scenarios for each produced char (Figure 1). Two material use scenarios were evaluated for the hydrochar: agricultural use (SH1) with potential substitution of fertilizers, and peat replacement in a growing media for horticulture (SH2). The energy utilization of the hydrochar was modeled as a mono-incineration process substituting energy from natural gas (SH3) and a co-incineration process in a lignite power plant (SH4). As for pyrolysis, three material utilizations of the biochar were investigated: agricultural use (SP1) with a substitution of fertilizers and lime, peat replacement in horticulture (SP2) and the cascade agricultural use, including biochar dosage in a biogas plant to boost anaerobic digestion and the subsequent utilization on agricultural land (SP3). For the biochar energy use, only co-combustion was energetically possible, which was modeled as co-incineration in a lignite-fired power plant (SP4), analogous to SH4. A detailed description of the processes for the utilization of the chars is presented in the Supplementary Materials (Figures S1-S8).

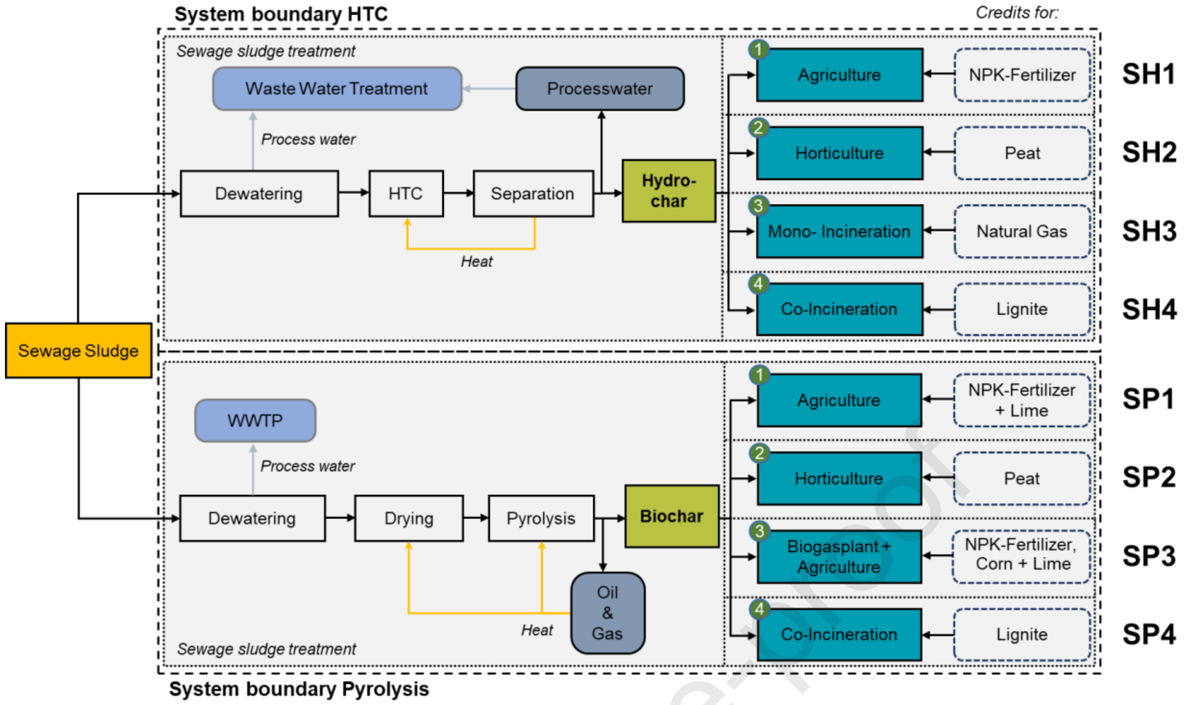

***Figure 1**: Simplified system boundaries of the modeled process chains of sewage sludge thermochemical treatment and subsequent biochar and hydrochar utilization in the HTC (SH1-SH4) and pyrolysis (SP1-SP4) scenarios [adapted from [41]].*

2.1.4 Geographical scope

The aim of the LCA was not to use detailed data from a single plant, but rather to derive generally applicable conclusions about different technologies and their potential emission behavior. Based on an evaluation at the national level, Germany served as the reference geographical framework. In the case of background data from the databases, the geographical scope of the aggregated processes is generally broader, as in some cases only European or global average processes were available. In order to extend the scope of the study in the sensitivity analysis, the conditions were later adapted to Swedish, Italian and Spanish contexts by adjusting the national electricity mixes.

2.1.5 Life cycle impact assessment (LCIA)

The impacts evaluated in the LCA included the influence on climate change, the eutrophication and acidification potential resulting from nutrients release into the environment, the stratospheric ozone depletion and ground-level ozone formation. Moreover, the human toxicity and ecotoxicity potentials were evaluated due to pollutants occurring in the process chains, as well as particulate matter emissions from combustion processes. Finally, the resource consumption was assessed due to the use of fossil fuels and raw materials. The ReCiPe 2016 method was selected to evaluate the overall environmental impacts [42]. The corresponding impact categories are shown in Table 1.

***Table 1**: Midpoint impact categories evaluated according to the ReCiPe methodology*

Life Cycle Impact Category	**Unit**
Global warming potential (GWP)	[kg CO_2-eq.]
Particulate matter formation potential (PM)	[kg $PM_{2,5}$-eq.]
Fossil fuel potential (FFP)	[kg oil-eq.]
Freshwater ecotoxicity potential (FETP)	[kg 1,4-DB-eq.]
Freshwater eutrophication potential (EP)	[kg P-eq.]
Ionising radiation potential (IRP)	[Bq. Co-60-eq.]
Ozone depletion potential (ODP)	[kg CFC-11-eq.]
Photochemical oxidant formation potential: ecosystems (OFP)	[kg NO_x-eq.]
Terrestrial acidification potential (AP)	[kg SO_2-eq.]
Terrestrial ecotoxicity potential (TETP)	[kg 1,4-DB-eq.]
Human toxicity potential, cancer (HTPc)	[kg 1,4-DB-eq.]
Human toxicity potential, non-cancer (HTPnc)	[kg 1,4-DB-eq.]

The potential long-term storage of the stable carbon amount contained in pyrolytic biochars offers an opportunity to remove carbon dioxide from the atmosphere, effectively counteracting global warming. One option for mapping the sequestered carbon quantities and their influence on the GWP in LCA would be not to include the quantity of sequestered CO_2-eq. emissions in the balance for the GWP, but to explicitly show this quantity separately or to relate the emissions to an amount of sequestered CO_2-eq. by using the functional unit [43,44]. However, since the aim of this work was to compare different waste treatment methods rather than the potential carbon sequestration performance, the proportion of carbon sequestration was integrated into LCIA to determine the GWP, aligning with current accounting practice [22,28,45–48]. The amount of sequestered CO_2 was calculated with the molar mass ratio of CO_2 and C (3.67 g g^{-1}), the biochar C content and a permanence coefficient of 0.65 for 100 years [49].

2.1.6 Data basis

As an inherent requirement for conducting a LCA, data is necessary for the material and energy flows of all the life cycle stages and underlying processes under consideration. The data is divided into foreground processes, which mainly represent the individual modeled process chain links, and background processes, which are based on generic data sets from the LCA databases. For this study, the GaBi and Ecoinvent (version 3.6) databases were used. However, due to the lack of practical data for the various investigated process chains, data from a variety of sources was used to model the foreground processes. Although the current literature provides useful information on the most important material and heat flows of the investigated technologies, the variance within data is significant and the overall data amount is limited, particularly concerning direct plant emissions [50]. Thus, industrial data from different plants, including research projects and personal communications, were integrated into the balancing to ensure the representation of the broadest possible spectrum of current technological situation.

2.2 Life cycle inventory (LCI)

The process chains were modeled according to the scenarios and system boundaries defined in Section 2.1. Before the digested sewage sludge is sent for thermochemical treatment, it is mechanically dewatered to a TS content of 20% using a screw press, as a common practice in wastewater treatment plants. The inventory of this process step and of the processes for downstream hydrochar and biochar utilization are described in the Supplementary Materials.

2.2.1 HTC

The digested and dewatered sewage sludge was considered as input material for the HTC process. The selected process conditions resulted from the consideration of a wide range of parameters from a diverse number of sources that have been analyzed in detail. On the one hand, the aim was to maximize the TS content while minimizing energy input. To achieve this, the selected process temperature should not be too high or too low and the treatment time should be adequate [4]. At the same time, good treatability of the resulting process water should be ensured by keeping the temperature as low and the treatment duration as short as possible to prevent the accumulation of refractory organic compounds and potential biological inhibitors [51]. On the other hand, the treatment should lead to a comprehensive removal of organic pollutants such as pharmaceutical residues, requiring the highest possible treatment temperature and duration. Following these preliminary considerations, the treatment temperature was set to 200 °C with a retention time of 1 hour. The modeling of the heat required for the specific material flow was initially based on mathematical considerations by Barber (2020) [52] (Equation 1). The results were then compared with industry and literature data published for similar substrate properties and plant configurations (Table S1), validated and adjusted whenever necessary (Table 2).

$$Q_{heat} = (c_{p_w} * m_w * \Delta T + c_{p_s} * m_s * \Delta T) \quad (1)$$

With: Q_{heat}= required amount of heat [kJ]; c_p= specific heat capacity [kJ kg^{-1} K^{-1}] of water (w) and solids (s); m= mass [kg] of water (w) and solids (s); ΔT= temperature difference between input and treatment temperature [K].

***Table 2**: Energy flows of the HTC process*

Parameter	Value	Unit	Source
Input			
Dewatered sewage sludge	1	kg	
Heat recovery	0.192	MJ	Calculated with data by [53]
External heat demand	0.467	-	Calculated
Exergy of the HTC process	98	%	Calculated with data by [53,54]
Electricity	0.072	MJ	[55,56]
Total heat demand	0.659	MJ	calculated
Output			
HTC Sludge	1	kg	
Heat losses	0.132	MJ	Calculated with data by [53,55–57]

After the HTC, the resulting hydrochar was separated using a chamber filter press, stored at the plant, transported and utilized in four different scenarios (SH1-SH4) (Figure 1).

2.2.2 Pyrolysis

The pyrolysis process was modeled using different plant parameters from pilot and industrial scale studies. Data from various pyrolysis plants, pilot projects and publications on biochar were used. The optimal treatment temperature and duration were set at 550 °C and two hours, respectively, to achieve the most complete removal of the organic pollutants while keeping a high phosphorus availability for crops [58–60]. Compared to HTC, the sewage sludge to be carbonized must be dried to at least 80% TS content before pyrolysis to optimize the process. For the start-up time of the pyrolysis reactor (2 hours) after weekly maintenance, the existing biogas boiler was used. The thermal energy required for start-up is 2.07 kJ kg^{-1} TS (sludge). Continuous operation of the pyrolysis plant requires 1.23 MJ kg^{-1} TS (sludge) of heat, which is provided by the combustion of the produced pyrolysis oils and gases. This amount of thermal energy includes the radiation and convection losses. To operate all ancillary units, 0.25 MJ kg^{-1} TS (sludge) of electrical energy is required.

Due to the heterogeneous and highly variable sewage sludge characteristics, it was difficult to obtain specific data for the modeled thermochemical conversion path with regard to the material and thermal properties of the resulting pyrolysis oils, gases and biochars. Nevertheless, an attempt was made to identify these core parameters for the selected process conditions based on literature data. The parameters selected for the modeled process (Table 3) were derived from a consolidated, quantitative analysis of various sources, from which the most plausible mean values were extracted and used for modeling [61–73]. The selected process conditions resulted in a mass-related proportion of products consisting of 51% biochar, 13% pyrolysis gas, and 36% pyrolysis oil (not condensed) in relation to the dry matter content of the input sludge. Potential gaseous emissions, which arise due to leaks in the pyrolysis reactor and process control [74,75], were allocated to the combustion process inventory. Following the pyrolysis process, the biochar was then stored at the plant, transported and utilized in four different scenarios (SP1-SP4).

Table 3*: Inventory of the pyrolysis process*

Parameter	Value	Unit	Source
Input			
Dried sewage sludge	1	kg	
Electrical energy	0.249	MJ kg^{-1} TS	[76]
Thermal energy	1.23	MJ kg^{-1} TS	Calculated after [76]
Thermal energy (start-up)	0.0021	MJ kg^{-1} TS	Calculated
Biochar share	51	%	[71,72]
Pyrolysis gas share	13	%	[71,72]
Pyrolysis oil share	36	%	[71,72]
Output			
Biochar	0.408	kg	Calculated
Pyrolysis gas	0.104	kg	Calculated
Pyrolysis oil (in gas phase)	0.288	kg	Calculated
Water vapor	0.2	kg	Calculated
Calorific value of biochar	8.6	MJ kg^{-1}	[70–72]
Calorific value of pyrolysis oil	13.8	MJ kg^{-1}	[71,72]
Calorific value of pyrolysis gas	15.8	MJ kg^{-1}	[71,72]

2.2.3 Biochar and hydrochar utilization scenarios

The use of the biochar and hydrochar produced by the two thermochemical conversion processes potentially offers various opportunities to mitigate the overall environmental burdens. For example, the long-term stable carbon storage through biochar soil application can counteract global warming and finite resources such as phosphorus can be recovered by closing material cycles with hydrochar and biochar. In addition, the energy use of the products can contribute to bioenergy production, thereby reducing reliance on fossil fuels. The selected scenarios were equally drawn up for both conversion processes to facilitate comparison (Figure 1). The detailed inventories of the processes in the various utilization scenarios can be found in the Supplementary Materials (Tables S5-S12).

2.3 Scenario and sensitivity analysis

Two methods were used to determine the sensitivity and uncertainty of the LCA model: first, a scenario analysis was performed, where different processes in the treatment chain and their background data were varied, considering the initial assumptions. For example, the thermal energy source (natural gas, electricity or biogas), or the electricity mix of the specific location may have a major influence on the emission characteristics of the overall system. The results from individual scenarios were then compared to the baseline to determine the model sensitivity to the individual input data. Additionally, alternative scenarios investigated the use of different flocculants, process water treatments (aerobic, anaerobic and wet oxidation), thermal energy supplies (biogas, natural gas, electricity) and electricity mixes (Sweden, Italy and Spain) for the thermochemical processes.

A sensitivity analysis was later performed through the variation of significant input parameters, examining their influence on the emission potentials in selected impact categories. In the sensitivity analysis, the identified parameters were varied within realistic bandwidths, considering the most recent literature data. The results within the various impact categories with parameter changes of ±10% (increased and decreased) of the initial value were evaluated and compared to the baseline. Parameters with an absolute result change of less than ±2.5% were classified as irrelevant, corresponding to an average model sensitivity to individual parameters [77]. The input parameters considered in the sensitivity analysis included the variation of the recovery rates of thermal energy within the treatment processes, the amount of flocculant used and peat to be substituted as well as the nutrient, heavy metal and carbon concentrations and the heating values of the produced hydrochar and biochar (Tables S17-S18).

Together, sensitivity and scenario analysis can support the validation of LCA results by highlighting data limitations and underpinning conclusions [35,78,79].

3 Results

3.1 Material flows within the treatment processes

The evaluation of the mass flows provides an initial reference point to assess the factors mostly influencing the emissions within the selected thermochemical conversion processes. The mass flows of both thermochemical processes show that a large fraction of the water initially contained in the sewage sludge can be separated by the first dewatering step, both for HTC and pyrolysis (see Figure 2). Efficient sludge dewatering is known to have a positive impact on downstream, typically energy-intensive, process steps, as higher dry substance contents enhance the overall efficiency and thus reduce the environmental impact and costs of the thermochemical processes [93,94]. In the specific case, the dewatered sewage sludge only accounts for around 25% of the initial amount of sludge from a mass balance perspective. In the HTC process chain, the dewatered sludge can be thermochemically treated directly, whereas pyrolysis requires prior drying. At the end of the process chain, 8.2% of the digested sludge from the HTC is available as hydrochar with a TS content of 60%, whereas for the pyrolysis, only 2.5% of the initial mass is converted into biochar, which is almost free of moisture.

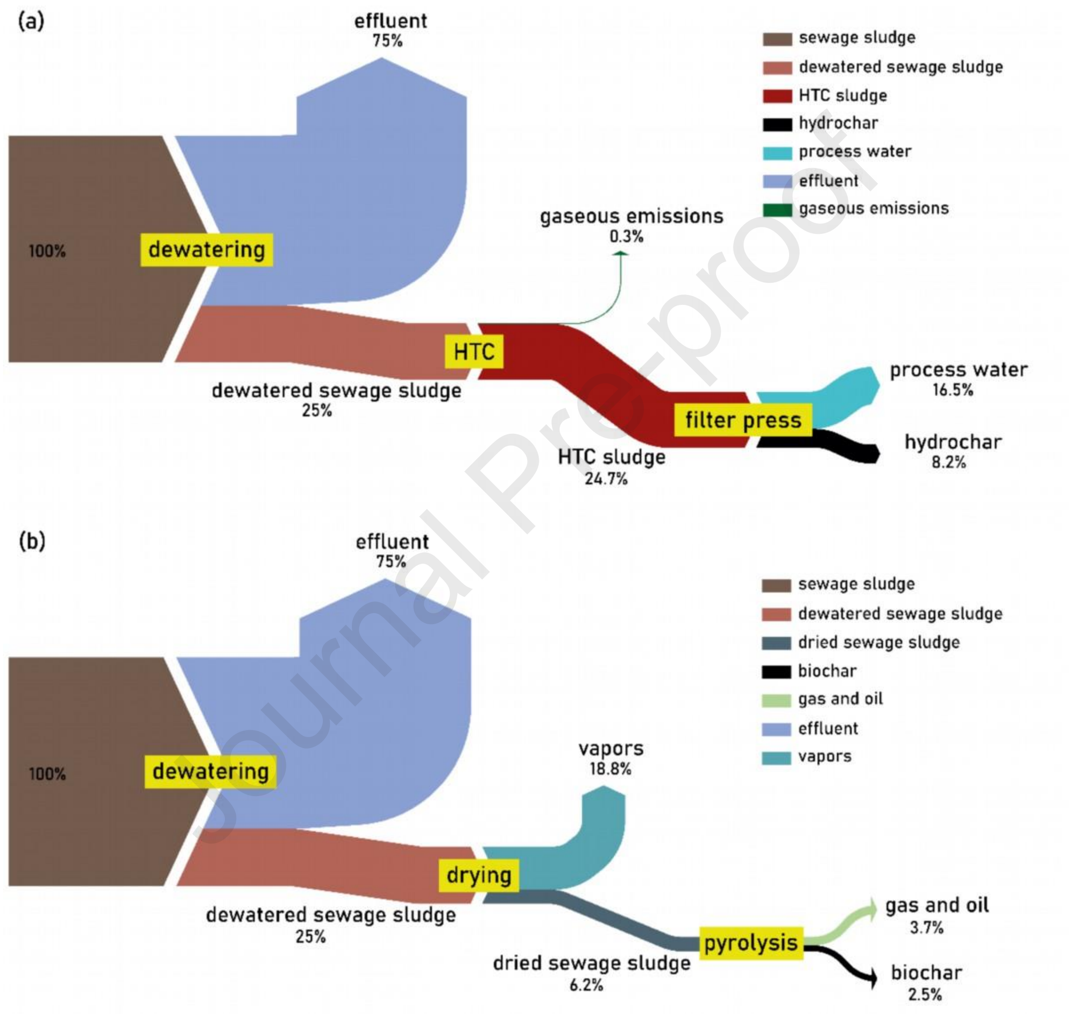

***Figure 2**: Mass flow of the modeled sewage sludge treatment by: (a) HTC and (b) pyrolysis [adapted from [41]]*

The evaluation of energy and mass flows indicates that the mass flows for biochar determined in this work align well with the literature values. Indeed, Ledakowicz et al. (2019) [69] report a biochar yield of only 1.75% for pyrolysis, but the pyrolysis process temperature selected for this work was lower (550 versus 800 °C), and higher temperatures are typically associated with lower biochar yields [4]. In an energy and material evaluation of process chains for the HTC of digested sewage sludge, Hämäläinen et al. (2021) [54] determined a similar hydrochar yield of 8% (50% TS) as derived in this work.

3.2 Environmental impacts of the HTC process chains

3.2.1 Hydrochar utilization scenarios

The GHG emissions from the HTC of sewage sludge are largely related to the provision of the energy required for the process, which leads to total emissions of 10.2 g CO_2-eq. FU^{-1} (Figure 3). The provision of thermal and electrical energy for operating the HTC alone causes 62% of the GWP (Figure S9).

The scenarios SH3-SH4 show that the greatest savings in GHG emissions are achieved by substituting fossil raw materials and energy, ranging from -71.4 g CO_2-eq. FU^{-1} (SH4) to -31.3 g CO_2-eq. FU^{-1} (SH1). In contrast, hydrochar use in agriculture (SH1) generates much lower credits from fertilizer substitution. The results indicate that the influence of the HTC treatment on the overall emissions is rather low. Carbonization in the aqueous medium saves the energy-intensive water evaporation that is required in pyrolysis processes and produces hydrochar with a high dry matter content. This reduces the energy required compared to drying and thus lowers GHG emissions. On the other hand, the credits for the substitution of fossil raw materials in the hydrochar application scenarios show that achieved emission savings largely depend on the reference scenarios with fossil-based background processes. Accordingly, the scenario for the substitution of energy from lignite (SH4) offers the highest emission reduction of FFP, while scenario SH1 exhibits the highest emissions overall.

For PM, the evaluation of the impact assessment shows that the emissions largely originate from the generic background process needed to provide the heat required for the HTC process. The credits for the energy from natural gas are minimal, as natural gas combustion generally releases lower particulate matter emissions than lignite (SH4). Therefore, particulate emissions are largely dependent on the selected form of substituted fossil resources. The results for the impact category EP show that the modeled phosphorus flows are mainly responsible for the occurring emissions. The biochar phosphorus content therefore not only leads to the desired fertilizing effect, but also contributes to a (desired) increase in the EP category in the material scenarios. In contrast to the EP, the evaluation for IRP shows that it is mainly influenced by selected electricity mix and, in the case of the biochar material application, the avoided emissions from fertilizer supply and peat extraction.

In toxicity potential assessments (FETP, TETP, HTPc and HTPnc), emissions mainly stem from polymer flocculant use and filtrate treatment prior to thermochemical conversion (Figure S9). In general, TETP shows higher emission potentials than FETP. The evaluation of the various scenarios regarding human toxicity highlights a predominant presence of organic pollutants such as polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) contained in the hydrochar for the carcinogenic effect fraction (HTPc). However, the elimination of pollutants through incineration contributes to emission reduction in scenarios SH3 and SH4.

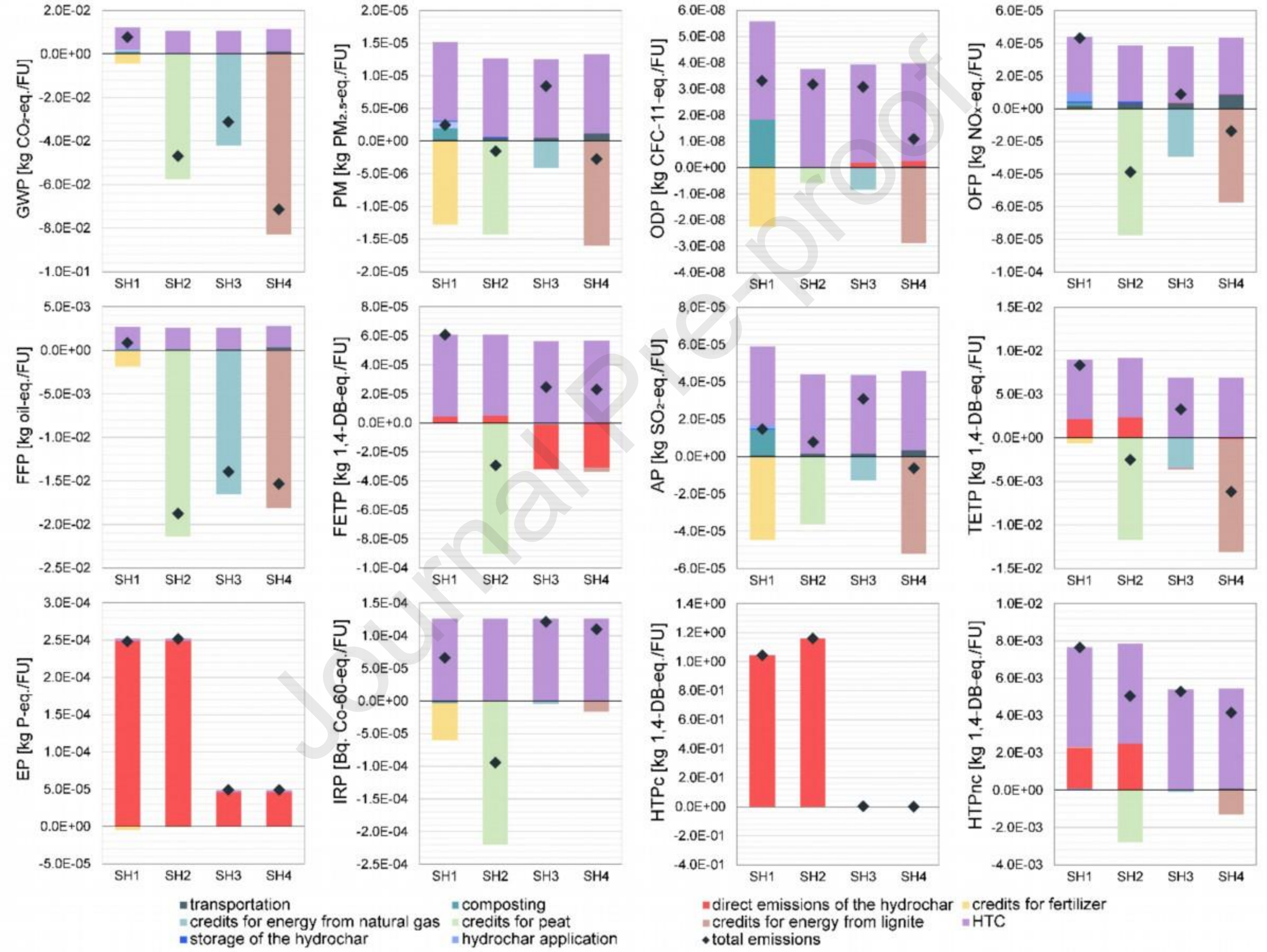

***Figure 3**: Environmental impacts of the different hydrochar utilization scenarios (SH1-SH4) for the HTC process chains [adapted from [41]].*

3.2.2 Scenario and sensitivity analysis

The LCIA results for the various process scenarios are illustrated in Figure 4, with dashed lines marking the results of the baseline scenario and bars representing the emissions resulting from the respective scenarios.

Replacing the polyacrylamide-based flocculant with the starch-based flocculant does not significantly alter the GWP, but leads to increased emissions in the impact categories PM, AP, EP, IRP, ODP and OFP, due to generally lower performance and higher chemical consumption. Compared to all other measures, the use of a starch-based flocculant shows the highest increase in the ecotoxicity (FETP and TETP) and human toxicity (HTPc and HTPnc). For anaerobic process water treatment, high emission savings are achieved in almost all impact categories, resulting in emission reductions. For wet oxidation treatment, instead, higher emissions are computed in the GWP, FFP and IRP categories due to the increased electricity requirements for oxidant production. For the PM, AP, EP, ODP and OFP categories, on the other hand, the integration of wet oxidation process water treatment leads to emission reductions compared to the baseline.

The shift of HTC's heat supply from biogas to natural gas reveals that in some impact categories the emissions decrease by up to 76% (ODP), whereas GWP and FFP increase by 37% and 108% respectively. Overall, the highest GWP increase of all scenarios occurs in the provision of the thermal energy required for HTC from electricity.

Transitioning to alternative European electricity mixes (Sweden, Spain, Italy) shows a decrease of GWP emissions by 9% (Italy) to 34% (Sweden), whereas the IRP emissions increase by 130% for Swedish conditions due to the high share of nuclear power. In the "best practice" scenario of HTC (Table S15), GWP is nearly halved (49%).

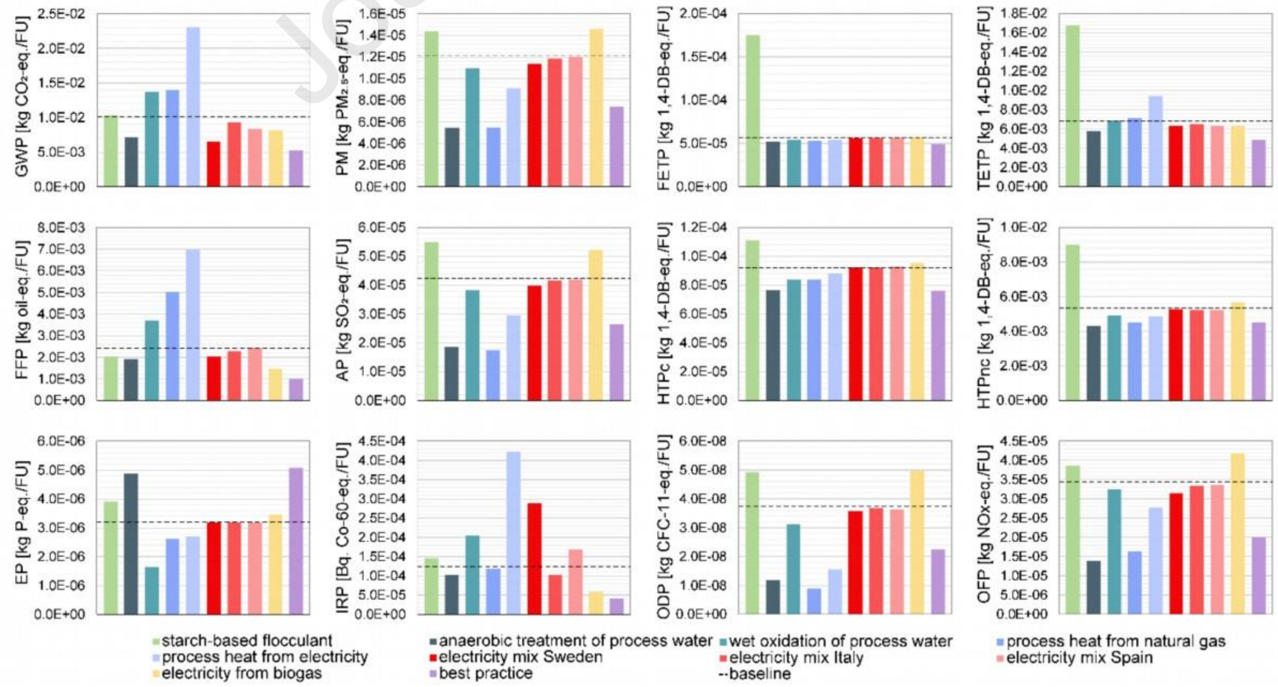

***Figure 4**: Environmental impact assessment of the various process adaptations for the HTC process chain [adapted from [41]].*

In contrast to the uncertainty associated with the HTC treatment, the model sensitivity to the hydrochar utilization scenarios reveals in some cases a high dependency on individual impact categories (Figure S11). However, in most cases, the highest and lowest emissions are attributable to the use of alternative process steps within thermochemical treatment rather than to specific parameter variations in the utilization, underscoring the overall dispersion of the results.

3.3 Environmental impacts of the pyrolysis process chains

3.3.1 Biochar utilization scenarios

The environmental impact of pyrolysis is assessed across the twelve selected environmental impact categories. Pyrolysis without biochar use yields a GWP of 0.2 kg CO_2-eq. FU^{-1} (Figure 5), whereby the largest shares are attributable to the generic background processes of providing thermal energy from biogas and electrical energy from the electricity mix for sludge drying (Figure S10). For the pyrolysis scenarios, the GWP shows that the two applications in agriculture have the highest emissions overall at 9.1 g CO_2-eq. FU^{-1} and 7.9 g CO_2-eq. FU^{-1} (Figure 5). Due to the credits for the substituted peat, biochar use in horticulture (SP2) generates emission savings of 8.8 g CO_2-eq. FU^{-1}. All three material uses (SP1-SP3) also offer the advantage that biochar use results in carbon sequestration, which is offset against GHG emissions with a negative emission of 8.1 g CO_2-eq. FU^{-1}. However, the highest savings and thus the lowest total emissions of -11.7 g CO_2-eq. FU^{-1} result in the scenario SP4, in which biochar use as an energy source replaces fossil energy production using lignite. Other ancillary processes in the individual scenarios, such as biochar transportation to the respective application sites, biochar application and incorporation to the fields as well as biochar direct emissions, only play a subordinate role for the GWP.

For PM, the results show that the largest proportion within the individual process chains results from the pyrolysis process itself. The credits in the agricultural scenarios (SP1 and SP3) are mainly caused by the avoidance of sulphur dioxide emissions in the provision of P fertilizer and, to a lesser extent, by direct dust emissions.

The FFP shows that the greatest savings can be achieved in scenarios in which biochar replaces fossil raw materials, whereas the majority of emissions in the FETP category comes from the upstream pyrolysis process, in which the production of the flocculant polymer accounts for a high emissions fraction. The EP indicates a clear dominance of direct phosphorus emissions from the biochar for all scenarios. Accordingly, the influence of the biochar phosphorus content and its leachability can be directly read from the EP, as observed by the material use of hydrochar.

The results of the impact assessment for AP, ODP, OFP and IRP underline that the decisive influence occurs during the pyrolysis process chain and thus it accounts for the largest share of the respective life cycle emissions for all scenarios. In the case of HTPnc, the heavy metal emissions from the application scenarios are decisive for the emission characteristics. Since the heavy metals from the biochar are not released into the environment during energy use, the toxicity potential in SP4 is the lowest compared to the material use scenarios. Interestingly, the direct heavy metal emissions from the biochar have less influence on the emission potential than the emissions from the background processes in all material utilization scenarios (SP1-SP3). The HTPc shows a clear dominance of a single material flow (very low residual amount of dioxins in the biochar after pyrolysis) which can be omitted through biochar incineration (SP4).

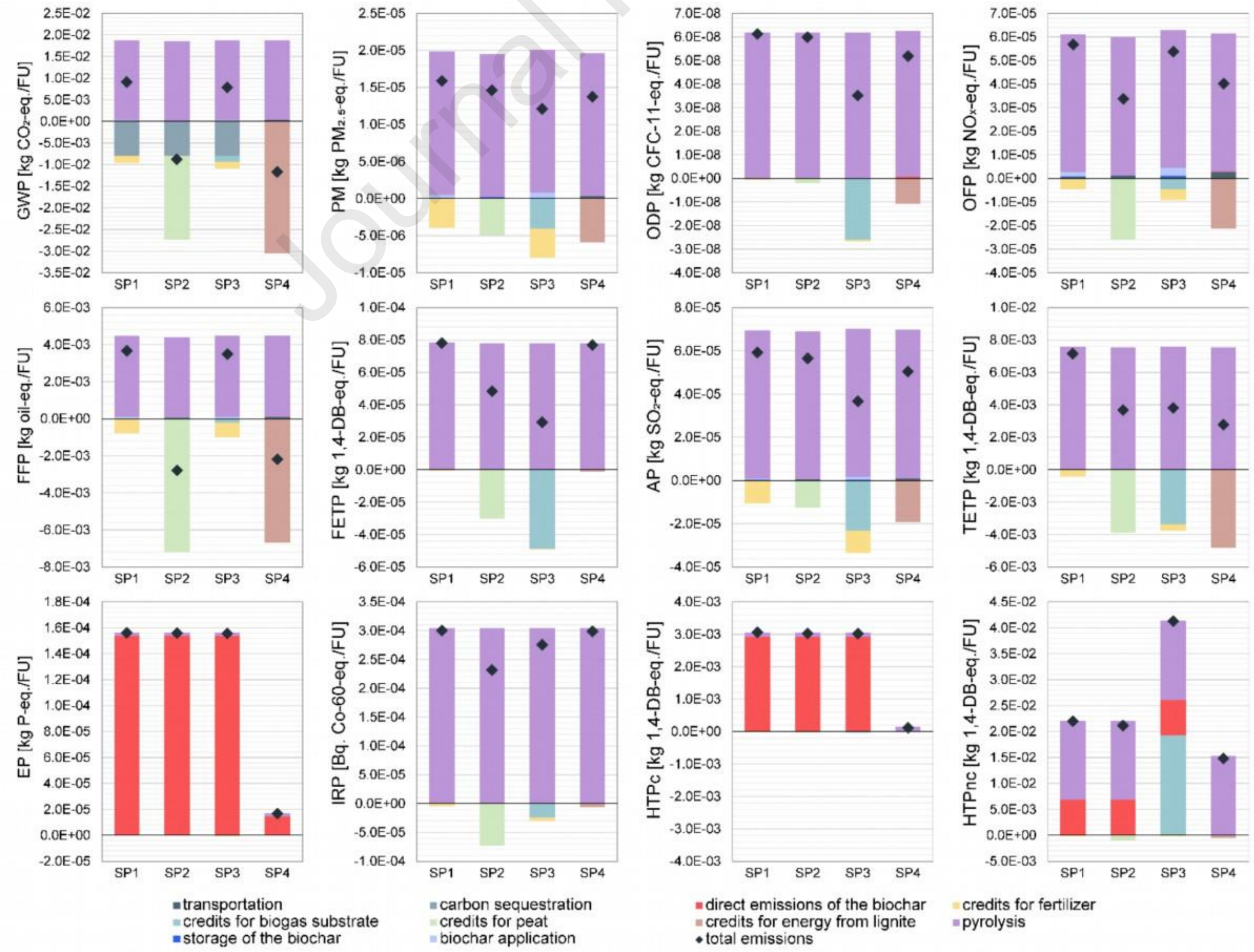

***Figure 5**: Environmental impacts of the different biochar utilization scenarios (SP1-SP4) for the pyrolysis process chains [adapted from [41]].*

3.3.2 Scenario and sensitivity analysis

The model sensitivity to individual plant configurations and future developments in energy supply is determined on the basis of the modification of individual processes within the process chain for sludge pyrolysis. The evaluation of model sensitivity to individual plant configurations reveals for the use of a starch-based flocculant no significant change in GWP, as shown in the sensitivity analysis of the HTC process (Figure 6). In the impact categories PM, AP, EP, IRP, ODP, FETP, TETP, HTP and OFP, the emissions increase, whereas fossil resource consumption (FFP) is reduced. Converting the heat supply to natural gas leads to an increase in emissions in the impact categories GWP (39%) and FFP (114%). If electrically operated heating elements are used to provide the energy required to operate the pyrolysis plant and the upstream drying process, emissions increase by 132% (GWP) to 197% (FFP), whereas for the remaining categories, emission reductions up to 67% (ODP) occur. The switch to other European electricity mixes (Sweden, Spain, Italy) results in emission reductions of 44% (Sweden) to 9% (Italy) for the GWP, whereas the IRP increases for a Swedish electricity mix by 108% and decreases for the Italian electricity mix by 41%. The results for the use of electrical energy from biogas to operate the overall system show that emission savings are achieved for the GWP, FFP and IRP, while an increase of 23% to 38% is observed in the other categories.

The parameter variations of the complete process chains (SP1-SP4) exhibit similar high sensitivities to HTC scenarios (SH1-SH4), particularly concerning the amount of substituted fossil-based resources (Figure S12).

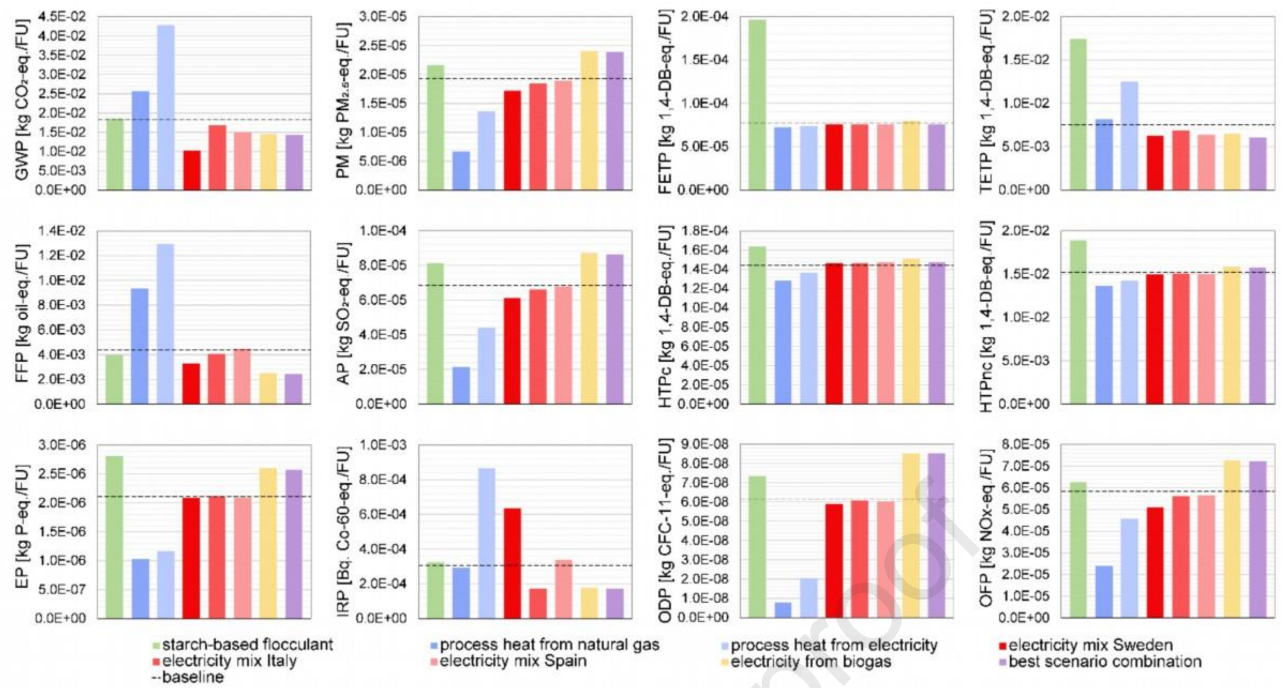

***Figure 6**: Environmental impact assessment of the various process adaptations for the pyrolysis process chain [adapted from [41]].*

3.4 Comparison of different utilization paths

The comparison of the various scenarios for hydrochar (SH1-SH4) and biochar utilization (SP1-SP4) along with the emission characteristics of the benchmark process is illustrated in Figure 7. Particularly in the impact categories of GWP and FFP, the pyrolysis scenarios produce higher emissions than HTC scenarios. However, the GHG emissions in all modeled scenarios are below those of the benchmark process and for the FFP only scenario SP1 and SP3 show a higher fossil resource consumption. In addition, all scenarios with substitution of fossil resources (SH2-SH4, SP2 and SP4) show emission reductions for both HTC and pyrolysis, whereby the credits for the substitution of fossil resources are higher for HTC due to the larger amount of hydrochar and substituted energy. For the scenarios of agricultural use of biochar and hydrochar, all process chains are at a similar level and show overall positive emissions. Therefore, the fertilizer substitution and the cascade use of biochar generate lower credits for the GWP than the substitution of fossil raw materials such as peat, natural gas and lignite. In addition, the optimal measures used in the sensitivity analysis, which characterize the lower end of the bandwidth, show that the emissions and credits almost balance each other out, giving thermochemical processes a clear advantage over mono-incineration. Therefore, both the pyrolysis and the HTC process chains have ecological advantages over mono-incineration for impact categories of GWP, PM, AP, ODP, IRP and OFP, regardless of whether the biochar or hydrochar is used for energy or material application. Overall, the HTC process chains show a clear advantage over the pyrolysis ones, mainly due to the elimination of the energy- and emission-intensive drying process. In addition, the comparison shows that there is no clear preference for the material or energy use of biochar, as different rankings in the emission characteristics occur both within the thermochemical processes and the impact categories, depending on the application scenario and the assumptions made.

When considering EP, the scenarios for the material use of biochar and hydrochar show a much higher impact potential than the scenarios for energy use due to the quantities of nutrients released into the environment. This emission potential is even higher for the HTC material scenarios (SH1 and SH2) than for the pyrolysis ones (SP1-SP3), as the plant availability of phosphorus in hydrochar is higher than in biochar and, in addition, during HTC part of the P is discharged with the process water.

The high toxicity potential in the HTPc category is attributed to the content of PCDD/Fs in hydrochar. While HTC treatment can somewhat reduce PCDD/Fs levels compared to raw sludge, specific HTC process conditions may exacerbate their toxicity potential [91,92]. The evaluation revealed that over 99% of the total toxicity potential is due to PCDDs emissions, while emissions of copper, cadmium, and zinc have a smaller impact when hydrochar is used as a material. In contrast, organic pollutants are largely eliminated during pyrolysis and incineration due to the higher process temperatures [58], resulting in much lower toxicity in these scenarios. For impact categories TETP, FETP and HTPnc, both thermochemical processes have lower emissions than the benchmark process in almost all scenarios However, the emission values can vary depending on the specific origin of the sewage sludge, with more contaminated sludge likely increasing the toxicity potential. In addition, the accumulation of persistent heavy metals could increase soil toxicity over decades, which is not reflected in the LCA study carried out.

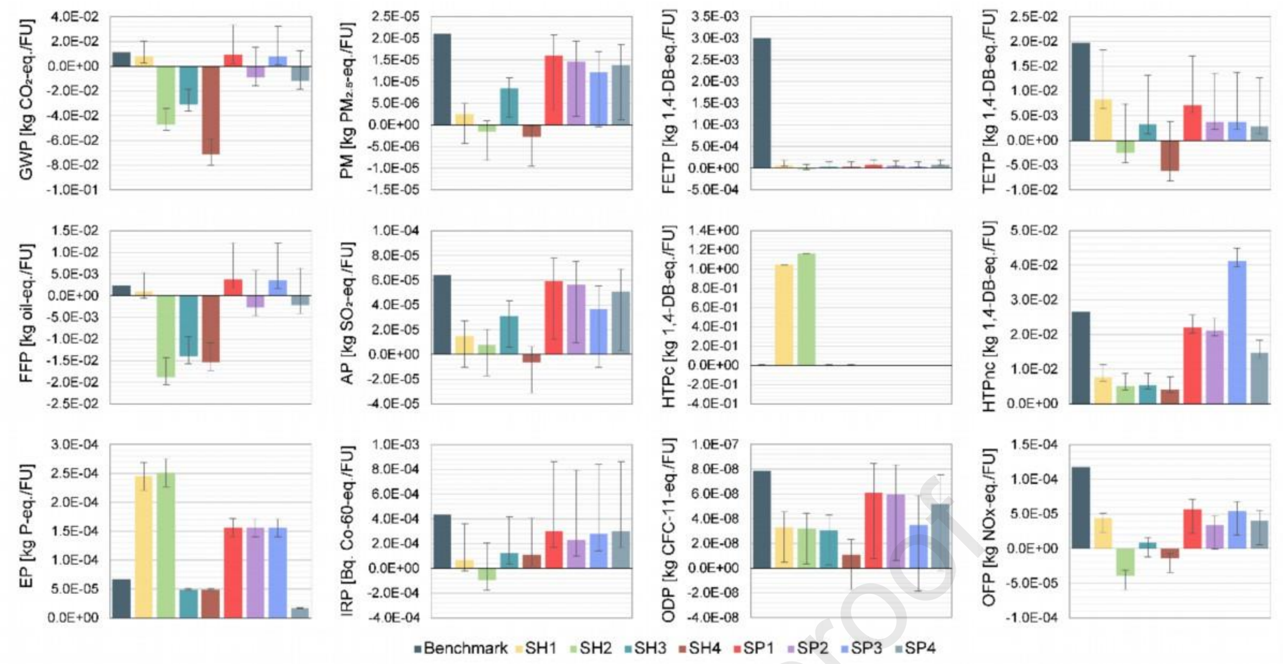

Figure 7*: Results of the LCIA of various process chains for the production and use of hydrochar (SH1-SH4) and biochar (SP1-SP4) compared to mono-incineration (benchmark). The whiskers reflect the maximum and minimum values of the sensitivity analysis for each scenario and impact category [adapted from [41]].*

4 Discussion

Due to the higher temperatures and therefore stricter process conditions of pyrolysis, a much larger proportion of the dry substance is transferred to the gas and liquid phase compared to HTC. In HTC, part of the organic matter ends up in the process water, which can also be used to generate energy through subsequent anaerobic digestion [54,80,81] and thus represents a valuable resource. In general, less biochar is obtained with pyrolysis compared to HTC. For this reason, and due to the higher calorific value of hydrochar compared to biochar, the credits that are achieved from the substitution of fossil energy are generally higher in the HTC than in the pyrolysis scenarios. Although pyrolysis produces by-products (pyrolysis oil and gas), that can directly provide part of the heat required for pyrolysis and sludge drying due to their high energy content, HTC integration as an alternative for sludge drying offers energy advantages, positively impacting the emission characteristics of the thermochemical process used (see Figure 3 and Figure 5 as well as Figure S9 and Figure S10). In addition, pyrolysis integration enables synergies within the occurring energy and material flows through the local provision and use of thermal energy embedded in the sludge. However, factors such as the local energy supply and its emissions demonstrate the influence of site-specific conditions on the overall environmental impact.

A limitation to consider when including credits from substituted fossil fuels through combustion processes is that the results of an LCA sometimes favour incineration over material recycling due to higher credits generated by replacing fossil energy [82]. However, through a fast transition towards renewable energies, material recycling may represent the better long-term alternative.

Moreover, although the credits from carbon sequestration are insufficient to offset the emissions generated in the process chains and the carbon sequestration potential of biochar produced from sewage sludge is generally low compared to other feedstocks [28], it can contribute to an effective carbon sink, especially in future defossilized energy systems.

Conducting a comprehensive comparison between different LCAs of pyrolysis or HTC, including the various (other) application pathways of the resulting chars, is challenging. Each researcher applies distinct methodologies, resulting in variations in functional units, allocations, impact categories and methods, thereby complicating comparisons [83]. Therefore, to thoroughly evaluate the impact of alternative sewage sludge processes, all specific HTC and pyrolysis parameters, as well as the selected LCIA methods, must be taken into account. This complexity makes discussing the current results in relation to the literature particularly complex and makes direct numerical comparisons nearly impossible.

However, in an LCA of various sewage sludge treatment methods, Tarpani et al. (2020) [84] determined that the highest GHG emissions of all investigated scenarios occurred in the pyrolysis scenario. This was based on sewage sludge drying using natural gas and biochar landfilling without crediting the carbon sequestration. In the impact categories related to toxicity assessment (FETP and HTP) as well as for PM and AP, however, pyrolysis achieved savings compared to incineration. Havukainen et al. (2022) [85] reached a similar conclusion, finding that co-pyrolysis of sewage sludge and wood waste caused higher emissions than sludge incineration, not only for the GWP, but also for the impact categories PM, TETP and EP.

Mayer et al. (2021) [11] conducted a LCA of HTC, pyrolysis and mono-incineration of digested sewage sludge, including the subsequent biochar incineration and P recovery from the ashes. Their results showed lower environmental impacts for HTC than for direct sludge application, whereas pyrolysis caused higher emissions than mono-incineration and direct agricultural sludge application. Analogously to the present results, the HTC process chain therefore showed the lowest emissions and the main reason for the high GWP of pyrolysis was the provision of the required amount of heat for sludge drying by natural gas. A high variance in the results was also observed across all impact categories. The highest emissions for the HTPnc resulted from mono-incineration, whereas the values for pyrolysis and HTC were comparatively low, consistently with the current results.

One reason for the wide variability of literature results is the definition of system boundaries. Although a LCA should always analyze the entire system from cradle to grave, the analyses often differ in terms of where the system begins and ends [86,87]. Other differences that make a direct comparison difficult are the technical assumptions made and the limited data availability resulting from a lack of full-scale experience for biochar and hydrochar production and utilization [2,85,88], mainly associated to legal hurdles [4,89].

Moreover, sewage sludge composition, which can vary greatly, has a major impact on the energy and pollutant content of the resulting chars and the energy flows for the treatments due to carbon and ash content, moisture level, and pollutants preloading. Therefore, even with the same treatment processes, LCA results can differ significantly [90]. The obtained results thus provide an initial assessment of the different recycling paths and show the differences between the possible advantages and disadvantages of material and energy use of the chars. In addition, due to the complex, interdependent parameter relationships and the generally high variability in the input materials, treatment parameters and final treatment products [79], the results are difficult to transfer to individual wastewater treatment plants.

In this study, the effects of plant construction and infrastructure creation were not taken into account, as high energy flows generally occur in thermochemical processes and therefore the influence of plant construction on environmental impacts is low. However, the smaller the plant and the less energy-intensive the process, the greater the influence of construction and demolition on the environmental impact [35].

In addition, allocation problems can arise in LCA for wastewater treatment paths and sewage sludge recycling [36], which can lead to different conclusions depending on the objective of the study. It is therefore a major challenge to transfer the results obtained to individual conditions. While HTC and pyrolysis are promising technologies, the technological maturity of these processes and legislative restrictions on the use of the products could still hinder their widespread adoption.

Nevertheless, the results show that the recovery of valuable materials and energy can have a positive environmental impact on sewage sludge treatment, whereby the extent of the benefit largely depends on the specific sludge characteristics, the various application scenarios and the different impact categories. For a comprehensive sustainability assessment, however, further studies should examine techno-economic and social aspects in addition to environmental impacts to evaluate the benefits and downsides of different treatment pathways for optimum sludge management.

5 Conclusions

HTC process chains generally show lower emissions than pyrolysis and both thermochemical treatments (HTC and pyrolysis) show lower emissions than the benchmark process of incineration in almost all impact categories. Moreover, biochar energy applications perform better in some categories than the material reuse in agriculture. The advantages of HTC and pyrolysis are even clearer when taking into account an optimum plant configuration.
The material use of hydrochar in agriculture and horticulture shows an increased toxicity potential due to the organic pollutants, which should be critically monitored to avoid soil contamination deriving from pre-polluted sewage sludge. This is particularly true for low-temperature conversion processes such as HTC, which do not completely remove harmful pollutants such as PCDD/PCDFs. Therefore, in addition to incineration, a further thermochemical treatment would be beneficial to reduce the negative effects on the toxicity potential due to organic contaminants. However, a combined or cascade treatment, where first HTC and then (high-temperature) pyrolysis is applied, poses an economic challenge that must be investigated in future studies. For other pollutants, no significant increases in toxicity potential were determined for either process. Although biochar can store carbon in the long term, the level of carbon storage was too low to compensate for the GHG emissions generated within the process chain. In addition, direct nutrient recycling is possible using the char from both thermochemical conversion processes, substituting mineral fertilizers. The results from the energy application of chars show that the credits achieved through the substitution of fossil supply for both HTC and pyrolysis offers ecological advantages in almost all environmental impact categories. With a properly tailored recycling strategy, both pyrolysis and HTC offer advantages over mono-incineration. Finally, in future defossilized energy systems, material recycling may be the better long-term alternative to energy use, if the pollutant content of the chars complies with the limits, thus continuous and careful material characterization will be mandatorily required. In addition, it is essential for the implementation of the proposed disposal paths that the legislative framework conditions must be adapted to enable these recycling paths accordingly.

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CRediT authorship contribution statement

Conceptualization: Fabian Gievers, Achim Loewen, Michael Nelles, Matia Mainardis and Arianna Catenacci; Methodology: Fabian Gievers; Formal analysis and investigation: Fabian Gievers; Writing - original draft preparation: Fabian Gievers, Matia Mainardis and Arianna Catenacci; Writing - review and editing: Michael Nelles and Achim Loewen; Visualization: Fabian Gievers; Supervision: Achim Loewen and Michael Nelles; Funding acquisition: Achim Loewen.

References

[1] R.R. Bora, R.E. Richardson, F. You, Resource recovery and waste-to-energy from wastewater sludge via thermochemical conversion technologies in support of circular economy: a comprehensive review, BMC Chemical Engineering 2 (2020) 8271. https://doi.org/10.1186/s42480-020-00031-3.

[2] A. Ding, R. Zhang, H.H. Ngo, X. He, J. Ma, J. Nan, G. Li, Life cycle assessment of sewage sludge treatment and disposal based on nutrient and energy recovery: A review, Science of The Total Environment 769 (2021) 144451. https://doi.org/10.1016/j.scitotenv.2020.144451.

[3] A. Amann, O. Zoboli, J. Krampe, H. Rechberger, M. Zessner, L. Egle, Environmental impacts of phosphorus recovery from municipal wastewater, Resources, Conservation and Recycling 130 (2018) 127–139. https://doi.org/10.1016/j.resconrec.2017.11.002.

[4] A. Catenacci, G. Boniardi, M. Mainardis, F. Gievers, G. Farru, F. Asunis, F. Malpei, D. Goi, G. Cappai, R. Canziani, Processes, applications and legislative framework for carbonized anaerobic digestate: Opportunities and bottlenecks. A critical review, Energy Conversion and Management 263 (2022) 115691. https://doi.org/10.1016/j.enconman.2022.115691.

[5] X. Meng, Q. Huang, J. Xu, H. Gao, J. Yan, A review of phosphorus recovery from different thermal treatment products of sewage sludge, Waste Disposal & Sustainable Energy 1 (2019) 99–115. https://doi.org/10.1007/s42768-019-00007-x.

[6] L. Egle, H. Rechberger, J. Krampe, M. Zessner, Phosphorus recovery from municipal wastewater: An integrated comparative technological, environmental and economic assessment of P recovery technologies, Science of The Total Environment 571 (2016) 522–542. https://doi.org/10.1016/j.scitotenv.2016.07.019.

[7] European Commission, Sewage Sludge, 2022. https://environment.ec.europa.eu/topics/waste-and-recycling/sewage-sludge_en (accessed 5 September 2022).

[8] W. Kwapinski, I. Kolinovic, J.J. Leahy, Sewage Sludge Thermal Treatment Technologies with a Focus on Phosphorus Recovery: A Review, Waste Biomass Valorization 12 (2021) 5837–5852. https://doi.org/10.1007/s12649-020-01280-2.
[9] E. Medina-Martos, I.-R. Istrate, J.A. Villamil, J.-L. Gálvez-Martos, J. Dufour, Á.F. Mohedano, Techno-economic and life cycle assessment of an integrated hydrothermal carbonization system for sewage sludge, Journal of Cleaner Production (2020) 122930. https://doi.org/10.1016/j.jclepro.2020.122930.
[10] S.G. Nkuna, T.O. Olwal, S.D. Chowdhury, J.M. Ndambuki, A review of wastewater sludge-to-energy generation focused on thermochemical technologies: An improved technological, economical and socio-environmental aspect, Cleaner Waste Systems 7 (2024) 100130. https://doi.org/10.1016/j.clwas.2024.100130.
[11] F. Mayer, R. Bhandari, S.A. Gäth, Life cycle assessment of prospective sewage sludge treatment paths in Germany, Journal of Environmental Management 290 (2021) 112557. https://doi.org/10.1016/j.jenvman.2021.112557.
[12] R. Canziani, G. Boniardi, A. Turolla, Phosphorus recovery—recent developments and case studies, in: Sustainable and Circular Management of Resources and Waste Towards a Green Deal, Elsevier, 2023, pp. 269–281. https://doi.org/10.1016/B978-0-323-95278-1.00007-3.
[13] M. Hušek, R. Homma, J. Moško, M. Pohorelý, K. Oshita, P-recovery versus current sewage sludge treatment policy in the Czech Republic and Japan, Clean Technologies and Environmental Policy 26 (2023) 1883–1899. https://doi.org/10.1007/s10098-023-02679-w.
[14] M. Schnell, T. Horst, P. Quicker, Thermal treatment of sewage sludge in Germany: A review, Journal of Environmental Management 263 (2020) 110367. https://doi.org/10.1016/j.jenvman.2020.110367.
[15] S. Tominc, V. Ducman, W. Wisniewski, T. Luukkonen, G.M. Kirkelund, L.M. Ottosen, Recovery of Phosphorus and Metals from the Ash of Sewage Sludge, Municipal Solid Waste, or Wood Biomass: A Review and Proposals for Further Use, Materials 16 (2023). https://doi.org/10.3390/ma16216948.
[16] G. Farru, F.B. Scheufele, D. Moloeznik Paniagua, F. Keller, C. Jeong, D. Basso, Business and Market Analysis of Hydrothermal Carbonization Process: Roadmap toward Implementation, Agronomy 14 (2024) 541. https://doi.org/10.3390/agronomy14030541.
[17] M. Cavali, N. Libardi Junior, R.d.A. Mohedano, P. Belli Filho, R.H.R. da Costa, A.B. de Castilhos Junior, Biochar and hydrochar in the context of anaerobic digestion for a circular approach: An overview, Science of Total Environment 822 (2022) 153614. https://doi.org/10.1016/j.scitotenv.2022.153614.
[18] M. Pimenta-Ocampo, G. Gascó, A. Méndez, W. Otten, R. Sakrabani, Pyrolysis or hydrothermal carbonisation for anaerobic-digested sewage sludge? A comparison of pyrochar and hydrochar structure and stability, Biomass Conversion and Biorefinery. (2023). https://doi.org/10.1007/s13399-023-05019-6.
[19] X. Li, R. Wang, C. Shao, D. Li, S. Bai, N. Hou, X. Zhao, Biochar and Hydrochar from Agricultural Residues for Soil Conditioning: Life Cycle Assessment and Microbially Mediated C and N Cycles, ACS Sustainable Chemistry & Engineering 10 (2022) 3574–3583. https://doi.org/10.1021/acssuschemeng.1c08074.
[20] T. Ekvall, G. Assefa, A. Björklund, O. Eriksson, G. Finnveden, What life-cycle assessment does and does not do in assessments of waste management, Waste Management 27 (2007) 989–996. https://doi.org/10.1016/j.wasman.2007.02.015.
[21] R. Salemdeeb, R. Saint, F. Pomponi, K. Pratt, M. Lenaghan, Beyond recycling: An LCA-based decision-support tool to accelerate Scotland's transition to a circular

economy, Resources, Conservation & Recycling Advances 13 (2022) 200069. https://doi.org/10.1016/j.rcradv.2022.200069.

[22] E.S. Azzi, E. Karltun, C. Sundberg, Life cycle assessment of urban uses of biochar and case study in Uppsala, Sweden, Biochar 4 (2022). https://doi.org/10.1007/s42773-022-00144-3.

[23] V. Bisinella, S. Schmidt, A.S. Varling, D. Laner, T.H. Christensen, Waste LCA and the future, Waste Management 174 (2024) 53–75. https://doi.org/10.1016/j.wasman.2023.11.021.

[24] L. Chen, Y. Zhang, C. Labianca, L. Wang, S. Ruan, C.S. Poon, Y.S. Ok, D.C.W. Tsang, Carbon-negative cement-bonded biochar particleboards, Biochar 4 (2022). https://doi.org/10.1007/s42773-022-00185-8.

[25] M.Z. Hauschild, R.K. Rosenbaum, S.I. Olsen (Eds.), Life Cycle Assessment: Theory and Practice, Springer International Publishing, Cham, 2018. https://doi.org/10.1007/978-3-319-56475-3.

[26] Z. Jiang, Y. Zou, Y. Li, F. Kong, D. Yang, Environmental life cycle assessment of supercapacitor electrode production using algae derived biochar aerogel, Biochar 3 (2021) 701–714. https://doi.org/10.1007/s42773-021-00122-1.

[27] C. Huang, B.A. Mohamed, L.Y. Li, Comparative life-cycle assessment of pyrolysis processes for producing bio-oil, biochar, and activated carbon from sewage sludge, Resources, Conservation and Recycling 181 (2022) 106273. https://doi.org/10.1016/j.resconrec.2022.106273.

[28] F. Xia, Z. Zhang, Q. Zhang, H. Huang, X. Zhao, Life cycle assessment of greenhouse gas emissions for various feedstocks-based biochars as soil amendment, Science of The Total Environment 911 (2024) 168734. https://doi.org/10.1016/j.scitotenv.2023.168734.

[29] G. Mannarino, S. Caffaz, R. Gori, L. Lombardi, Environmental Life Cycle Assessment of Hydrothermal Carbonization of Sewage Sludge and Its Products Valorization Pathways, Waste and Biomass Valorization (2022) 1–20. https://doi.org/10.1007/s12649-022-01821-x.

[30] A. Ogunleye, J. Flora, N. Berge, Life Cycle Assessment of Hydrothermal Carbonization: A Review of Product Valorization Pathways, Agronomy 14 (2024) 243. https://doi.org/10.3390/agronomy14020243.

[31] A. Hosseinian, P. Brancoli, N. Vali, J. Ylä-Mella, A. Pettersson, E. Pongrácz, Life cycle assessment of sewage sludge treatment: Comparison of pyrolysis with traditional methods in two Swedish municipalities, Journal of Cleaner Production 455 (2024) 142375. https://doi.org/10.1016/j.jclepro.2024.142375.

[32] ISO, 14040:2006 Environmental management — Life cycle assessment — Principles and framework. International Organization for Standardization, 2006.

[33] M. Brandão, M. Martin, A. Cowie, L. Hamelin, A. Zamagni, Consequential Life Cycle Assessment: What, How, and Why?, in: M. Brandão (Ed.), Encyclopedia of Sustainable Technologies: Consequential Life Cycle Assessment: What, How, and Why?, Elsevier, 2017, pp. 277–284. https://doi.org/10.1016/B978-0-12-409548-9.10068-5.

[34] ISO, 14044:2006 Environmental management — Life cycle assessment — Requirements and Guidelines. International Organization for Standardization, 2006.

[35] L. Corominas, D.M. Byrne, J.S. Guest, A. Hospido, P. Roux, A. Shaw, M.D. Short, The application of life cycle assessment (LCA) to wastewater treatment: A best practice guide and critical review, Water Research 184 (2020) 116058. https://doi.org/10.1016/j.watres.2020.116058.

[36] S. Heimersson, M. Svanström, T. Ekvall, Opportunities of consequential and attributional modelling in life cycle assessment of wastewater and sludge

management, Journal of Cleaner Production 222 (2019) 242–251. https://doi.org/10.1016/j.jclepro.2019.02.248.

[37] Joint Research Centre, International Reference Life Cycle Data System (ILCD) Handbook: General guide for life cycle assessment: detailed guidance, Publications Office of the European Union, Luxembourg, 2010. https://doi.org/10.2788/38479.

[38] T.H. Christensen, A. Damgaard, J. Levis, Y. Zhao, A. Björklund, U. Arena, M.A. Barlaz, V. Starostina, A. Boldrin, T.F. Astrup, V. Bisinella, Application of LCA modelling in integrated waste management, Waste Management 118 (2020) 313–322. https://doi.org/10.1016/j.wasman.2020.08.034.

[39] M. Pradel, L. Aissani, J. Villot, J.-C. Baudez, V. Laforest, From waste to added value product: towards a paradigm shift in life cycle assessment applied to wastewater sludge – a review, Journal of Cleaner Production 131 (2016) 60–75. https://doi.org/10.1016/j.jclepro.2016.05.076.

[40] L. Lombardi, C. Nocita, E. Bettazzi, D. Fibbi, E. Carnevale, Environmental comparison of alternative treatments for sewage sludge: An Italian case study, Waste Management 69 (2017) 365–376. https://doi.org/10.1016/j.wasman.2017.08.040.

[41] F. Gievers, Vergleichende Untersuchungen und Bilanzierungen von Prozessketten zur Herstellung und Nutzung von Biokohlen aus Klärschlämmen. Dissertation, University of Rostock, 2023. https://doi.org/10.18453/rosdok_id00004455.

[42] M.A.J. Huijbregts, Z.J.N. Steinmann, P.M.F. Elshout, G. Stam, F. Verones, M. Vieira, M. Zijp, A. Hollander, R. van Zelm, ReCiPe2016: A harmonised life cycle impact assessment method at midpoint and endpoint level, International Journal of Life Cycle Assessment 22 (2017) 138–147. https://doi.org/10.1007/s11367-016-1246-y.

[43] P. Goglio, A.G. Williams, N. Balta-Ozkan, N. Harris, P. Williamson, D. Huisingh, Z. Zhang, M. Tavoni, Advances and challenges of life cycle assessment (LCA) of greenhouse gas removal technologies to fight climate changes, Journal of Cleaner Production 244 (2020) 118896. https://doi.org/10.1016/j.jclepro.2019.118896.

[44] T. Terlouw, C. Bauer, L. Rosa, M. Mazzotti, Life cycle assessment of carbon dioxide removal technologies: a critical review, Energy & Environmental Science 14 (2021) 1701–1721. https://doi.org/10.1039/D0EE03757E.

[45] E.S. Azzi, E. Karltun, C. Sundberg, Small-scale biochar production on Swedish farms: A model for estimating potential, variability, and environmental performance, Journal of Cleaner Production 280 (2021) 124873. https://doi.org/10.1016/j.jclepro.2020.124873.

[46] J. Cooper, L. Dubey, A. Hawkes, Life cycle assessment of negative emission technologies for effectiveness in carbon sequestration, Procedia CIRP 105 (2022) 357–361. https://doi.org/10.1016/j.procir.2022.02.059.

[47] D. Lefebvre, A. Williams, G.J. Kirk, J. Meersmans, S. Sohi, P. Goglio, P. Smith, An anticipatory life cycle assessment of the use of biochar from sugarcane residues as a greenhouse gas removal technology, Journal of Cleaner Production 312 (2021) 127764. https://doi.org/10.1016/j.jclepro.2021.127764.

[48] J. Matuštík, M. Pohorelý, V. Kočí, Is application of biochar to soil really carbon negative? The effect of methodological decisions in Life Cycle Assessment, Science of The Total Environment 807 (2022) 151058. https://doi.org/10.1016/j.scitotenv.2021.151058.

[49] D. Woolf, J. Lehmann, S. Ogle, A.W. Kishimoto-Mo, B. McConkey, J. Baldock, Greenhouse Gas Inventory Model for Biochar Additions to Soil, Environmental Science & Technology 55 (2021) 14795–14805. https://doi.org/10.1021/acs.est.1c02425.

[50] H. Chang, Y. Zhao, S. Zhao, A. Damgaard, T.H. Christensen, Review of inventory data for the thermal treatment of sewage sludge, Waste Management 146 (2022) 106–118. https://doi.org/10.1016/j.wasman.2022.05.002.
[51] J.D. Marin-Batista, A.F. Mohedano, J.J. Rodríguez, M.A. de La Rubia, Energy and phosphorous recovery through hydrothermal carbonization of digested sewage sludge, Waste Management 105 (2020) 566–574. https://doi.org/10.1016/j.wasman.2020.03.004.
[52] W. Barber, Sludge Thermal Hydrolysis: Application and Potential, IWA Publishing, Ashland, 2020. https://doi.org/10.2166/9781789060287.
[53] C. Remy, J. Stüber, Weiterentwicklung des Klima- und Ressourceneffizienzpotentials durch HTC-Behandlung ausgewählter Berliner Klärschlämme – HTC-Berlin (2015).
[54] A. Hämäläinen, M. Kokko, V. Kinnunen, T. Hilli, J. Rintala, Hydrothermal carbonisation of mechanically dewatered digested sewage sludge-Energy and nutrient recovery in centralised biogas plant, Water Research 201 (2021) 117284. https://doi.org/10.1016/j.watres.2021.117284.
[55] M. Buttmann, TerraNova® Ultra Sludge Drying and Phosphorous Recovery by Ultradewatering of Sludge, in: 2nd International Symposium on Hydrothermal Carbonization | Hydrothermal carbonization as a building block for a sustainable bioeconomy | May, 14th – 16th 2019, Berlin, Leipzig: DBFZ, 2019.
[56] A. Kuhles, Hydrothermal Carbonization and Biogas – Synergy effects. Example of the HTC Innovation campus Rheinmuehle, in: 2nd International Symposium on Hydrothermal Carbonization | Hydrothermal carbonization as a building block for a sustainable bioeconomy | May, 14th – 16th 2019 | Berlin, Leipzig: DBFZ, 2019.
[57] M. Owsianiak, M.W. Ryberg, M. Renz, M. Hitzl, M.Z. Hauschild, Environmental Performance of Hydrothermal Carbonization of Four Wet Biomass Waste Streams at Industry-Relevant Scales, ACS Sustainable Chemistry & Engineering (2016). https://doi.org/10.1021/acssuschemeng.6b01732.
[58] W. Buss, Pyrolysis Solves the Issue of Organic Contaminants in Sewage Sludge while Retaining Carbon—Making the Case for Sewage Sludge Treatment via Pyrolysis, ACS Sustainable Chemistry & Engineering 9 (2021) 10048–10053. https://doi.org/10.1021/acssuschemeng.1c03651.
[59] C.C. de Figueiredo, A.d.S.P.J. Reis, A.S. de Araujo, L.E.B. Blum, K. Shah, J. Paz-Ferreiro, Assessing the potential of sewage sludge-derived biochar as a novel phosphorus fertilizer: Influence of extractant solutions and pyrolysis temperatures, Waste Management 124 (2021) 144–153. https://doi.org/10.1016/j.wasman.2021.01.044.
[60] F. Yu, J. Wang, X. Wang, Y. Wang, Q. Guo, Z. Wang, X. Cui, Y. Hu, B. Yan, G. Chen, Phosphorus-enriched biochar from biogas residue of Eichhornia crassipes: transformation and release of phosphorus, Biochar 5 (2023). https://doi.org/10.1007/s42773-023-00281-3.
[61] D. Barry, C. Barbiero, C. Briens, F. Berruti, Pyrolysis as an economical and ecological treatment option for municipal sewage sludge, Biomass and Bioenergy 122 (2019) 472–480. https://doi.org/10.1016/j.biombioe.2019.01.041.
[62] M. Breulmann, M. van Afferden, R.A. Müller, E. Schulz, C. Fühner, Process conditions of pyrolysis and hydrothermal carbonization affect the potential of sewage sludge for soil carbon sequestration and amelioration, Journal of Analytical and Applied Pyrolysis 124 (2017) 256–265. https://doi.org/10.1016/j.jaap.2017.01.026.
[63] V. Frišták, M. Pipíška, G. Soja, Pyrolysis treatment of sewage sludge: A promising way to produce phosphorus fertilizer, Journal of Cleaner Production 172 (2018) 1772–1778. https://doi.org/10.1016/j.jclepro.2017.12.015.

[64] N. Gao, K. Kamran, C. Quan, P.T. Williams, Thermochemical conversion of sewage sludge: A critical review, Progress in Energy and Combustion Science 79 (2020) 100843. https://doi.org/10.1016/j.pecs.2020.100843.
[65] G. Gerasimov, V. Khaskhachikh, O. Potapov, G. Dvoskin, V. Kornileva, L. Dudkina, Pyrolysis of sewage sludge by solid heat carrier, Waste Management 87 (2019) 218–227. https://doi.org/10.1016/j.wasman.2019.02.016.
[66] P.K. Ghodke, A.K. Sharma, J.K. Pandey, W.-H. Chen, A. Patel, V. Ashokkumar, Pyrolysis of sewage sludge for sustainable biofuels and value-added biochar production, Journal of Environmental Management 298 (2021) 113450. https://doi.org/10.1016/j.jenvman.2021.113450.
[67] M. Hušek, J. Moško, M. Pohořelý, Sewage sludge treatment methods and P-recovery possibilities: Current state-of-the-art, Journal of Environmental Management 315 (2022) 115090. https://doi.org/10.1016/j.jenvman.2022.115090.
[68] C. Karaca, S. Sözen, D. Orhon, H. Okutan, High temperature pyrolysis of sewage sludge as a sustainable process for energy recovery, Waste Management 78 (2018) 217–226. https://doi.org/10.1016/j.wasman.2018.05.034.
[69] S. Ledakowicz, P. Stolarek, A. Malinowski, O. Lepez, Thermochemical treatment of sewage sludge by integration of drying and pyrolysis/autogasification, Renewable and Sustainable Energy Reviews 104 (2019) 319–327. https://doi.org/10.1016/j.rser.2019.01.018.
[70] Z. Liu, S. Singer, Y. Tong, L. Kimbell, E. Anderson, M. Hughes, D. Zitomer, P. McNamara, Characteristics and applications of biochars derived from wastewater solids, Renewable and Sustainable Energy Reviews 90 (2018) 650–664. https://doi.org/10.1016/j.rser.2018.02.040.
[71] J. Moško, M. Pohorelý, S. Skoblia, Z. Beňo, M. Jeremiáš, Detailed Analysis of Sewage Sludge Pyrolysis Gas: Effect of Pyrolysis Temperature, Energies 13 (2020) 4087. https://doi.org/10.3390/en13164087.
[72] M. Tomasi Morgano, H. Leibold, F. Richter, D. Stapf, H. Seifert, Screw pyrolysis technology for sewage sludge treatment, Waste Management 73 (2018) 487–495. https://doi.org/10.1016/j.wasman.2017.05.049.
[73] A.B.H. Trabelsi, K. Zaafouri, A. Friaa, S. Abidi, S. Naoui, F. Jamaaoui, Municipal sewage sludge energetic conversion as a tool for environmental sustainability: production of innovative biofuels and biochar, Environmental Science and Pollution Research 28 (2021) 9777–9791. https://doi.org/10.1007/s11356-020-11400-z.
[74] G. Cornelissen, N.R. Pandit, P. Taylor, B.H. Pandit, M. Sparrevik, H.P. Schmidt, Emissions and Char Quality of Flame-Curtain “Kon Tiki” Kilns for Farmer-Scale Charcoal/Biochar Production, PLoS One 11 (2016) e0154617. https://doi.org/10.1371/journal.pone.0154617.
[75] J.F. Peters, D. Iribarren, J. Dufour, Biomass Pyrolysis for Biochar or Energy Applications?: A Life Cycle Assessment, Environmental Science & Technology 49 (2015) 5195–5202. https://doi.org/10.1021/es5060786.
[76] Pyreg, sewage sludge, 2020. https://dev.pyreg.de/wp-content/uploads/2020_PYREG_flyer_sludge_EN.pdf (accessed 2 January 2022).
[77] J. Cohen, P. Cohen, S.G. West, Aiken. L.S., Applied Multiple Regression/Correlation Analysis for the Behavioral Sciences, Routledge, New York, 2002. https://doi.org/10.4324/9780203774441.
[78] M. Niero, M. Pizzol, H.G. Bruun, M. Thomsen, Comparative life cycle assessment of wastewater treatment in Denmark including sensitivity and uncertainty analysis, Journal of Cleaner Production 68 (2014) 25–35. https://doi.org/10.1016/j.jclepro.2013.12.051.

[79] X. Zhou, J. Li, X. Zhao, J. Yang, H. Sun, S.-S. Yang, S. Bai, Resource recovery in life cycle assessment of sludge treatment: Contribution, sensitivity, and uncertainty, Science of The Total Environment 806 (2022) 150409. https://doi.org/10.1016/j.scitotenv.2021.150409.

[80] J.D. Marin-Batista, J.A. Villamil, J.J. Rodriguez, A.F. Mohedano, M.A. de La Rubia, Valorization of microalgal biomass by hydrothermal carbonization and anaerobic digestion, Bioresource Technology 274 (2019) 395–402. https://doi.org/10.1016/j.biortech.2018.11.103.

[81] K. Meisel, A. Clemens, C. Fühner, M. Breulmann, S. Majer, D. Thrän, Comparative Life Cycle Assessment of HTC Concepts Valorizing Sewage Sludge for Energetic and Agricultural Use, Energies 12 (2019) 786. https://doi.org/10.3390/en12050786.

[82] T. Ekvall, M. Gottfridsson, M. Nellström, J. Nilsson, M. Rydberg, T. Rydberg, Modelling incineration for more accurate comparisons to recycling in PEF and LCA, Waste Management 136 (2021) 153–161. https://doi.org/10.1016/j.wasman.2021.09.036.

[83] S. Elfallah, A. Benzaouak, O. Bayssi, A. Hirt, A. Mouaky, H. El Fadili, S. Rachidi, E.M. Lotfi, N. Touach, M. El Mahi, Life cycle assessment of biomass conversion through fast pyrolysis: A systematic review on technical potential and drawbacks, Bioresource Technology Reports 26 (2024) 101832. https://doi.org/10.1016/j.biteb.2024.101832.

[84] R.R.Z. Tarpani, C. Alfonsín, A. Hospido, A. Azapagic, Life cycle environmental impacts of sewage sludge treatment methods for resource recovery considering ecotoxicity of heavy metals and pharmaceutical and personal care products, Journal of Environmental Management 260 (2020) 109643. https://doi.org/10.1016/j.jenvman.2019.109643.

[85] J. Havukainen, A. Saud, T.F. Astrup, P. Peltola, M. Horttanainen, Environmental performance of dewatered sewage sludge digestate utilization based on life cycle assessment, Waste Management 137 (2022) 210–221. https://doi.org/10.1016/j.wasman.2021.11.005.

[86] J. Matuštík, T. Hnátková, V. Kočí, Life cycle assessment of biochar-to-soil systems: A review, Journal of Cleaner Production 259 (2020) 120998. https://doi.org/10.1016/j.jclepro.2020.120998.

[87] J. Moreno, J.J. Espada, Environmental and techno-economic assessment of thermochemical treatment systems for sludge, in: Wastewater Treatment Residues as Resources for Biorefinery Products and Biofuels, Elsevier, 2020, pp. 201–223. https://doi.org/10.1016/B978-0-12-816204-0.00010-2.

[88] J. Huang, Y. Feng, H. Xie, P. Wu, M. Wang, B. Wang, Q. Zhang, S. Zhang, Z. Liu, A bibliographic study reviewing the last decade of hydrochar in environmental application: history, status quo, and trending research paths, Biochar 5 (2023). https://doi.org/10.1007/s42773-023-00210-4.

[89] J. Paz-Ferreiro, A. Nieto, A. Méndez, M.P.J. Askeland, G. Gascó, Biochar from Biosolids Pyrolysis: A Review, International Journal of Environmental Research and Public Health 15 (2018). https://doi.org/10.3390/ijerph15050956.

[90] S.K. Teoh, L.Y. Li, Feasibility of alternative sewage sludge treatment methods from a lifecycle assessment (LCA) perspective, Journal of Cleaner Production 247 (2020) 119495. https://doi.org/10.1016/j.jclepro.2019.119495.

[91] H. Brookman, F. Gievers, V. Zelinski, J. Ohlert, A. Loewen, Influence of Hydrothermal Carbonization on Composition, Formation and Elimination of Biphenyls, Dioxins and Furans in Sewage Sludge, Energies 11 (2018) 1582. https://doi.org/10.3390/en11061582.

[92] G. Liberatori, C. Mazzoli, F. Ferraro, L. Sturba, M.L. Vannuccini, D. Baroni, P.A. Behnisch, M. Puccini, S. Vitolo, I. Corsi, Aryl hydrocarbon reporter gene bioassay for

screening polyhalogenated dibenzo-p-dioxins/furans and dioxin-like polychlorinated biphenyls in hydrochar and sewage sludge, Journal of Hazardous Materials 428 (2022) 128256. https://doi.org/10.1016/j.jhazmat.2022.128256.

[93] Z. Liu, R. Balasubramanian, Upgrading of waste biomass by hydrothermal carbonization (HTC) and low temperature pyrolysis (LTP): A comparative evaluation, Applied Energy 114 (2014) 857–864. https://doi.org/10.1016/j.apenergy.2013.06.027.

[94] H. Luo, F. Cheng, B. Yu, L. Hu, J. Zhang, X. Qu, H. Yang, Z. Luo, Full-scale municipal sludge pyrolysis in China: Design fundamentals, environmental and economic assessments, and future perspectives, Sci. Total Environ. 795 (2021) 148832. https://doi.org/10.1016/j.scitotenv.2021.148832.

Highlights:

- HTC has energy advantages over pyrolysis and therefore lower emissions
- The material use of the chars enables nutrient recycling and carbon sequestration
- HTC and pyrolysis chains have a lower environmental impact than mono-incineration

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: