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Environmental performance of hydrothermal carbonization of four wet biomass waste streams at industry-relevant scales

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Abstract

Hydrothermal carbonization (HTC) of green waste, food waste, organic fraction of municipal solid waste (MSW), and digestate is assessed using life cycle assessment as a potential technology to treat biowaste. Water content of the biowaste and composition of the resulting hydrochar are important parameters influencing environmental performance. Hydrochar produced from green waste performs best and second best in respectively 2 and 10 out of 15 impact categories, including climate change, mainly due to low transportation needs of the biowaste and optimized pumping efficiency for the feedstock. By contrast, hydrochar produced from the organic fraction of MSW performs best in 6 impact categories, but has high potential impacts on human health and ecosystems caused by emissions of toxic elements through ash disposal. Greatest potential for environmental optimization for the HTC technology is in the use of heat and electricity with increasing plant size, but its overall environmental performance is largely influenced in a given geographic location by the incumbent waste management system that it replaces. Impact scores are within range of existing alternative treatment options, suggesting that despite being relatively immature technology, and depending on the geographic location of the plant, HTC may be an attractive treatment option for biowaste.

Keywords

bioenergy, biowaste, hydrochar, life cycle assessment, upscaling

1. Introduction

Hydrothermal carbonization (HTC) is attracting attention as an environmental technology to treat biowaste, including municipal solid waste, while producing the carbonaceous material hydrochar.[1–7] However, the environmental performance of the HTC at scales relevant to industry, considering the need for separate biowaste collection and post-treatment of the resulting hydrochar, has to date, not been reported in the literature. Here, we report on a life cycle assessment (LCA) of a pilot- and full commercial-scale HTC plant which has been carried out to identify the processes in the underlying life cycle with the largest potential for optimization, and ultimately to support the environmentally conscious design of future HTC plants.

During HTC, biomass is dehydrated in the presence of water by applying temperature (around 180-250 °C) and pressure (around 10-20 bars).[1,8] The main products of HTC are the carbonaceous material hydrochar, process water containing various inorganic and organic compounds, and non-condensable gases.[9,10] Hydrochar has properties that make it a good candidate for use as solid fuel, soil conditioner with carbon storage value, or a material for construction of battery electrodes.[11–18] HTC plants are based on either vertical (e.g. AVA-CO2, TerraNova Energy GmbH) or horizontal reactors (e.g. Artec Biotechnologie GmbH, Grenol GmbH) in various configurations. The Spanish small-medium enterprise (SME) Ingelia S.L., has erected one of the first pilot-scale HTC plants, that employs one cylindrical vertical reactor operating continuously.[9] Wet biomass is fed from the bottom, the resulting hydrochar/water slurry is removed (also from the bottom) while the gases are collected from the top. To increase capacity, the SME plans to add a second reactor, and furthermore, more two- and four-reactor plants (with larger reactors relative to the pilot-scale reactor) will be installed in a near future in other countries. Other HTC technology developers also allow for upscaling of their plants and offer modular design of HTC installations (e.g. AVA-CO2).

Environmental performance of HTC is expected to change when upscaling to the full commercial-scale is done.[19–21] Table S1 of the SI†, Section S1, shows the potential implications of upscaling on environmental performance of HTC of biowaste. For example, higher input of steel, metals and crude oil per unit of plant is expected to cause linear increase of the impacts on climate change, resource depletion, and various toxicity- and non-toxicity related impact categories due to the need for manufacturing of additional reactors and plant equipment. Antagonistically, non-linear capacity increase as dimensions or reactors change and plant grows (resulting in higher hydrochar output per unit of plant) is expected to decrease these impacts, depending on the contribution of the plant materials to total life cycle impacts. Thus, an assessment of environmental performance of the technology must also consider the effects of size and capacities of the plant. Further, the environmental performance of HTC is expected to be influenced by the regular waste management system that HTC replaces.[22] Neither pilot- nor full commercial-scale performance of HTC considering these factors has, to date, been assessed using LCA.

Earlier efforts to characterize environmental performance of HTC are limited to one recent study by Berge et al.,[23] who showed how life cycle impacts of HTC of food-waste and combustion of the resulting hydrochar in a power-plant depend on process water emissions and the type of energy that is substituted. For example, emissions of metallic elements stemming from discharge of HTC process water drove toxic impacts on human health and ecosystems, while across all life cycle impact categories substituting energy derived from fossil sources, like anthracite or lignite resulted in the best environmental performance. Although their study highlighted the role of energy source that the hydrochar replaces, it has four limitations. First, Berge et al.[23] used lab scale data when parametrizing their model. Second, combustion of hydrochar (derived from food waste) was assumed to mimic that incineration of municipal solid waste (MSW). Third, they omitted several relevant impact categories from their assessment, including human health impacts from particulate matter (PM) and resource depletion. Finally, important processes were omitted from system boundaries, including: (i) separate biowaste collection, (ii) consumption of electricity for pumping of wet feedstock into the reactor and drying and pelletizing of the resulting hydrochar; and (iii) disposal of post-treatment and post-combustion ashes.

In this paper, we address these four limitations, and concurrently present life cycle inventory (LCI) and life cycle impact assessment (LCIA) results of HTC of green waste (being garden trimmings), food waste (represented by orange peels), organic fraction of municipal solid waste and digestate at industry-relevant scales. All waste-streams are promising candidates for hydrothermal carbonization at full commercial-scale as validated by a pilot-scale assessment. Primary data from pilot-plant operations were used to model the foreground processes. Emissions of CO2, CO, nitrous oxides (NOx), SO2 and particulate matter (PM) from hydrochar combustion were based on measurements, while emissions of metals were taken from generic ecoinvent process for incineration of biowaste while correcting for differences in composition and properties between hydrochar and biowaste. To illustrate the potential of the technology, environmental performance at pilot-scale with one reactor was compared to that at full commercial scale with two or four reactors. Full-scale plants differ from the pilot-scale mainly with regard to plant capacity (increasing capacity with increasing scale, resulting from increasing the number and dimensions of reactors) and energy and material inputs (decreasing inputs per treated quantity with increasing scale).

2. Methods

2.1. Wet biomass waste streams

Green waste is composed of herbaceous biomass (forest litter, leaves, flowers, fruits, seeds, twigs and woody material), collected separately as garden trimmings. This biowaste stream is a significant contributor to organic waste generation worldwide.[24] Food waste is represented in our study by orange peels. It is estimated that global citrus peel waste production is around 60-100 million tons a year.[25–27] The organic fraction of municipal solid waste (MSW), which is waste that has been separated from metals and plastics at the collection point, is a mixed biowaste sources that remains a global challenge.[28] Availability of digestate remaining after anaerobic digestion of agricultural biomass (that has been concentrated at the biogas plant prior collection) varies, and is the largest in regions where domination treatment option for agricultural waste is anaerobic digestion. All biowaste types were collected as separate fractions, including orange peels which are waste from juice making factory. Organic fraction of MSW was separated from other MSW fractions at the composting plant. Details of the incumbent waste management systems for each biowaste type are presented in Table 1. The composition of the waste streams influences the hydrochar properties with regard to emissions of particulate matter and metals during combustion, and release of metals from ash disposal. Here, the four streams differ in the content of water, nitrogen (N), sulfur (S), and ash. Compounding this is the fact that considerable heterogeneity exists within the composition of the ash itself. Distribution of metal between solid and liquid phases of the HTC slurry has earlier been identified as an important parameter determining the environmental performance of hydrochar derived from food waste,[23] but quantitative life-cycle based comparison taking into account biowaste-specific distribution between solid and liquid phases has, to date, not been reported in open literature.

2.2. Life cycle assessment

The LCA was conducted in accordance with the requirements of the ISO standard and the guidelines of the International Reference Life Cycle Data System (ILCD) handbook.[29,30]

Functional unit. Because the main function of hydrochar used as solid fuel is to release energy, the functional unit was defined as "*output of 1 MJ of heat to a building from a domestic 5-15 kW stove*".

System boundaries. Spain was chosen as the primary geographical scope of the assessment because this is where the pilot plant is located. The hydrochar pellets are transported from Spain to the UK, where they are sold as solid fuel for use in domestic heating. This is an ongoing business activity and a realistic scenario for the future; the British partner already distributes around 1 million tons of domestic solid fuels. With nearly 300 million tons of hard coal being used as solid fuel in Europe only, the potential of hydrochar as solid fuel are even larger.[31] The system boundaries included the whole underlying life cycle, from the construction of the HTC plant, post-treatment equipment and the stove, collection of biowaste and its conversion to raw hydrochar, removal of the ash using flotation, drying and pelletizing, transportation of hydrochar pellets and combustion in the stove, and finally decommissioning of the HTC plant and the stove (Fig. 1). Wood is combusted in a boiler at the plant to generate heat needed for running the HTC process, with a fraction used for drying cleaned hydrochar. HTC process water is concentrated using reverse osmosis, brought to citrus plantation, where it is diluted to reduce concentrations of metals, and used as fertilizer in agriculture. This is also an ongoing business activity.

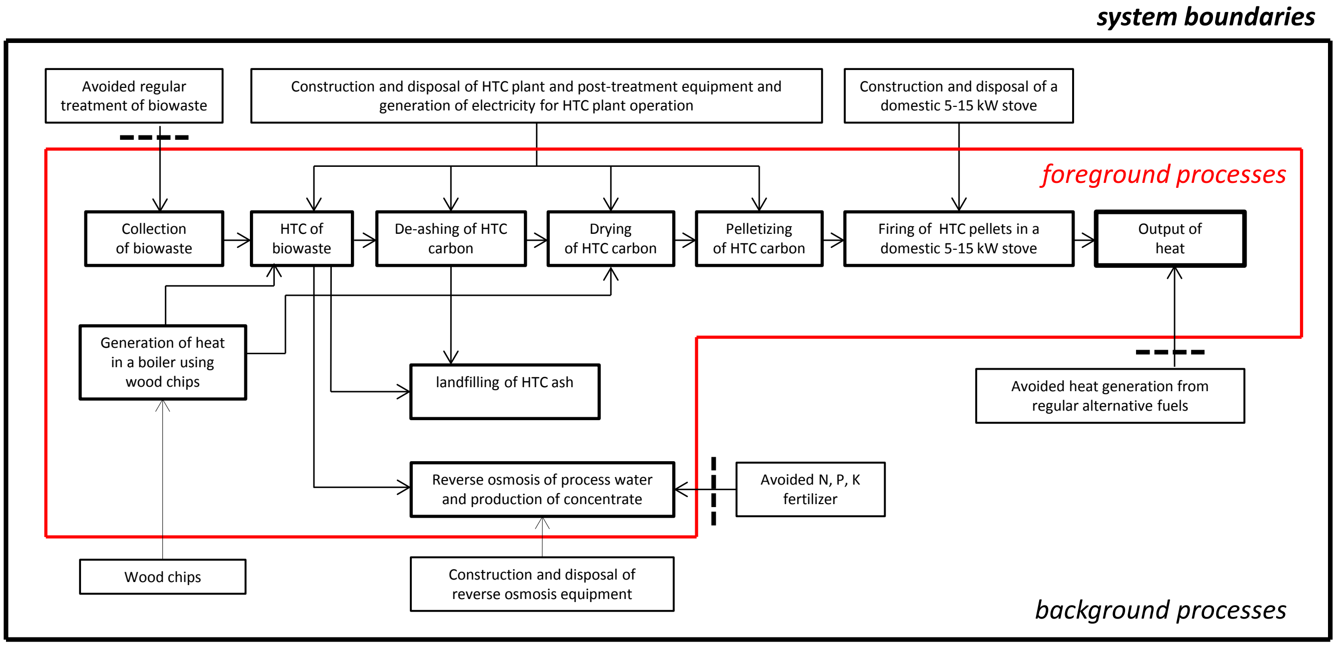

Fig. 1. System boundaries for hydrothermal carbonization of biowaste with energy recovery, with functional unit defined as “output of 1 MJ of heat to a building from a domestic 5-15 kW stove”. Dashed lines indicate avoided processes.

Sensitivity scenarios. To illustrate sensitivities of the LCA results to geographic location, a comparison was made for hydrochar produced and used as solid fuel in Germany, which is one of the largest potential users of carbonaceous products in Europe. Compared to Spain, this leads to differences in the modeling of collection of biowaste, generation of electricity, extraction of fossil coal, and conventional waste management system. In summary, sensitivity analysis considered differences in: (i) biowaste type; (ii) geographic location for the production and use of hydrochar; (iii) plant scale; and (iv) replaced waste management system (as determined by the geographic location of the production of hydrochar). Berge et al.[23] already studied the influence of substituted energy source, and hence, this was thus not considered here. Table 1 presents an overview of all 16 sensitivity scenarios.

Table 1. Overview of the compared scenarios for hydrothermal carbonization (HTC) wet biomass waste streams.

# Scenario	Sensitivity parameter	Geographic location (production/use) [a]	Biowaste type [c]	Transportation distance of the biowaste to the plant (in km)	Plant scale [b]	Replaced waste management system (WMS) [d]
1	Baseline	ES/UK	GW	7	Pilot, 1 reactor	COM
2-4	Biowaste type	ES/UK	FW; OFMSW; DG	7; 26; 36; 70	Pilot, 1 reactor	COM and INC (DG only)
5-12	Transportation distance of the biowaste to the plant (in km)	ES/UK	FW; OFMSW; DG	7; 7; 7; 7	Pilot, 1 reactor	COM and INC (DG only)
13-16	Plant scale	ES/UK	GW; FW; OFMSW; DG	7; 26; 36; 70	Full, 2 reactors; Full, 4 reactors	COM and INC (DG only)
17-20	Replaced waste management system (WMS)	DE/DE	GW; FW; OFMSW; DG	7; 26; 36; 70	Full, 4 reactors	INC (GW and DG) and COM (FW and OFMSW)

[a] ES: Spain; UK: the United Kingdom; DE: Germany

[b] at full commercial-scale the following parameters are different compared with the pilot-scale configuration: overall plant capacity, material inputs for construction of the HTC plant and the post-treatment equipment, heat input for running the HTC process, and

electricity use for pumping, drying and pelletizing (please see Table S3 for details of the model parameters at pilot- and full commercial-scale)

[c] GW: green waste, FW: food waste, OFMSW: organic fraction of municipal solid waste, DG: digestate

[d] COM: composting with fertilizer replacement, INC: incineration with energy recovery. Replaced waste management systems are based on the data retrieved from Eurostat for wood waste and vegetal waste categories, assumed to be representative of treatment of green waste and food waste, respectively in the concerned country. The dominant treatment options for wood waste are "recovery other than energy recovery" in Spain (97.7% of total wood waste) which we model as composting with fertilizer replacement, and "incineration with energy recovery" in Germany (76.2% of total wood waste). The dominant treatment option for vegetal waste in both Spain and Germany is "recovery other than energy recovery" (87.7 and 91.2% of total vegetal waste, respectively) which we also model as composting with fertilizer replacement. The organic fraction of MSW does not exist as a separate waste category in Eurostat and is also modelled as composting with fertilizer recovery, whereas digestate is expected to be incinerated with energy recovery.

Modeling framework. The ILCD guidelines provide methodological guidance according to different decision situations. The current study is in this context considered a micro-level decision support (type-A) situation since the production and use of hydrochar as solid fuel are not expected to cause structural changes on the market (e.g. decommissioning of existing waste management installations), at least at the current state of maturity and spread of the HTC technology. Therefore, the assessment applies an attributional approach, using average Spanish (or German) data and energy mixes and modelling average biowaste collection in the appropriate countries. Globally produced and traded commodities such as raw metals and alloys are modeled as global production, while the HTC plant and post-treatment equipment are modeled for European conditions. In cases of processes with recovery of commodities, system expansion was performed, where recycled steel substitutes the production of virgin steel, and that the process water concentrate substitutes production of inorganic fertilizers. Likewise, credits are given to avoided extraction and firing of fossil hard coal, and to avoided conventional treatment of biowaste in accordance with the recommendations of the ILCD guidelines for this decision support type.

Life cycle impact assessment. The product systems were modeled in SimaPro, version 8.0.4.30 (PRé Consultants bv, the Netherlands). Environmental impact scores were calculated using the ILCD's recommended practice characterization factors at midpoint (ILCD 2011 Midpoint+, version 1.05), as implemented in SimaPro.[32] This recommended practice has been identified by assessing a total of 156 different characterization models belonging to 12 different LCIA methodologies.[32] All ILCD impact categories were considered, apart from ionizing radiation impacts on ecosystems which considered not sufficiently representative for this type of impact. Ranking of biowaste streams may be sensitive to the inclusion/exclusion of long-term emissions (that is, emissions occurring after 100 years) which may determine the magnitude of eutrophication- and toxicity-related impact scores. Since the long-term emissions have larger uncertainties than short-term emissions, it is of interest to see how their inclusion affects the conclusions. Thus, the impact scores were calculated with long-term emissions either included (default settings) or excluded from the assessment. Normalization was done using the European set of ILCD's normalization factors for reference year 2010, version 4.0, as implemented in SimaPro. Synthesis of the LCIA methods and normalization factors are presented in SI†, Section S2.

2.3. Data and model parameters

Unit processes for the foreground system were constructed using model parameters based on measurements performed at a pilot plant at Ingelia S.L. (Valencia, Spain). They are synthesized in the SI†, Section S3. Background information of the plant itself is given elsewhere.[9] We measured parameters related to: (i) composition of the biowaste (i.e., content of water, ash, nitrogen (N), carbon (C), sulfur (S)); (ii) HTC plant and post-treatment equipment (i.e., material inputs, plant utilization rate, overall plant capacity, electricity and heat use for pumping of feedstock, electricity use for drying and pelletizing, yield of raw hydrochar and yield of hydrochar pellets, amount of process water, amount of gases); (iii) properties of hydrochar (i.e., content of water, ash, N, C, S, fluoride, chloride, and higher heating value of hydrochar pellets); (iv) combustion of hydrochar pellets (i.e., emissions of CO_2, CO particulate matter (PM); and (v) composition of the ash (phosphor (P), boron (B), and 19 metals and metalloids), composition of process water (N, P, B, and 19 metals and metalloids); and composition of gases (CO_2, CO, H_2). Emissions of PM, CO_2, CO, nitrogen oxides (NO_x), and SO_2 from hydrochar combustion in the stove are based on measurements performed during experiments using a pilot-scale (180 kW) grate combustion unit. Emissions of metallic elements to air were calculated using transfer coefficients for emissions to air from the ecoinvent process for incineration of biowaste, corrected for differences in composition and moisture between the hydrochar and the biowaste in the ecoinvent process. Life times of HTC plant, post-treatment equipment, reverse osmosis membrane, and buildings, and thermal efficiency of the boiler, were assumed using values based on reasonable expectations. Transportation distances between the plant, retail, and final user were taken from Google maps, whereas location of the final user (in the UK) is unknown and had to be assumed.

The parameters for the full commercial-scale process are estimated from the pilot plant values using scaling factors that consider optimization of the plant (e.g. reduction in heat and electricity inputs) and increased material needs, as presented by the technology developers in the business plan for a full commercial-scale plant in two- or four-reactor settings (see SI†, Section S3 for details). When upscaling from pilot to the full commercial-scale with two reactors, material input for the HTC plant increases by a factor of 2.2 when the number of reactors doubles. Reactors are of the same type owing to modular design, but the scaling factor is larger than 2 because dimensions of the reactors increase. At full commercial-scale both types and dimensions of reactors are the same and material input increases by a factor of 2 when number of reactors doubles. Material input for the post-treatment equipment increases by a factor of 1.7 when the number of reactors doubles, irrespective of the plant scale, since increasing dimensions of the post-treatment equipment rather than increasing the number of individual elements is most likely. We assumed no change in product quality with an increase in plant scale owing to the same types of HTC reactors and the same process conditions (temperature, heat).

Data for background processes, like construction and decommissioning of the HTC plant and the stove, or (avoided) production of inorganic fertilizers are based on generic processes available in ecoinvent, version 3.1.[33,34] Avoided waste treatment processes (like composting or incineration) were adapted to account for differences in biowaste water content, composition (such as content of carbon, nitrogen, metals, etc.) and properties (like degradability) between generic biowaste used in ecoinvent processes and the biowaste types considered in this study. Details of the adaptation of biowaste treatment processes are presented in the SI†, Section S3.

2.4. Uncertainty analysis

Uncertainties in the life cycle inventories for the foreground processes (e.g. in material inputs or emissions) were estimated using the Pedigree matrix approach.[35] Briefly, each uncertain data point was first assessed based on five data quality criteria (i.e. reliability, completeness, temporal correlation, geographical correlation, and further technological correlation) and corresponding uncertainty factors were assigned. Next, they were combined with a basic uncertainty factor (that depends on the type of data) and geometric standard deviations for the uncertain data point calculated, assuming that log-normal distribution applies to the data as uncertainty in processes often follows a skewed distribution.[36] Section S5 in the SI presents uncertainty factors and the squared geometric standard deviations for the foreground processes. Uncertainties in the background processes were based on geometric standard deviations already assigned to flows in the ecoinvent processes.

Monte Carlo simulations (1000 iterations) were carried out to compare the sensitivity scenarios while keeping track of the correlations between uncertainties of the compared systems. The employed modeling software only supports this when long-term impacts are included, and hence a statistical comparison between the scenarios was performed using long-term impacts. Comparison results were considered statistically significant if at least 95% of all 1000 Monte Carlo runs were favorable for one scenario.

3. Results and discussion

In the below, we address applicability of our life cycle inventories, illustrate general trends in LCIA results and present results for three selected impact categories. Then, we interpret our results and provide recommendations to technology developers on where to focus when optimizing the environmental performance of the technology. Finally, we address applicability of our findings in the biowaste management context.

3.1. Life cycle inventories

Unit processes and life cycle inventory (LCI) results are documented in the SI†, Section S4. They include all input and output flows from each unit process along the life cycle of the HTC. The inventory data is representative for plants developed based on HTC process running at Ingelia S.L., but LCA practitioners can readily adapt our unit processes to other HTC installations in future studies. Results presented in this paper will guide LCA practitioners about which processes are salient when using our LCI in future studies.

3.2. Overview of life cycle impact assessment results

Figure 2 shows characterized life cycle impact assessment (LCIA) results for the baseline scenario and the scenario showing the influence of plant scale for four selected impact categories. They represent typical impact profiles observed for the four wet biomass waste streams. The results scores for other scenarios across all 15 ILCD’s impact categories are presented in SI†, Section S6 (Tables S39-S47). The LCIA results show four main trends. First, the impact scores are negative for 6 (green waste, food waste, and digestate) and 5 (organic fraction of MSW) impact categories (Table S39 and S40). s Third, green waste is seen as the best or second best in 2 and 10 impact categories, respectively, and statistically significant differences in impact scores between green waste and food waste, organic fraction of MSW, and digestate occur in 14, 12, and 6 impact categories, respectively. Fourth, digestate is seen the worst in 7 impact categories although in 10 impact categories the difference between digestate and food waste is not statistically significant (Table S39 and S40).

Normalized results show that across all waste streams and impact categories, negative impact scores are the lowest for the impact categories climate change, human toxicity, non-cancer (apart from the organic fraction of MSW where impact scores are positive), particulate matter, and acidification, where they are below 0.1% of the annual impact of an average European (see SI†, Section S6, Fig. S1 and S2). Positive impact scores are in the same range and the highest for resource depletion, freshwater ecotoxicity, and human toxicity (cancer and non-cancer, for hydrochar derived from organic fraction of MSW). Weighing factors are not yet available for ILCD methods, but assuming an equal weight across impact categories, processes and emissions contributing to these seven impact categories are the primary drivers of the environmental performance of hydrothermal carbonization.

Ranking of biowaste streams in these three impact categories changes when long-term impacts are excluded. Exclusion of long-term impacts is the most important for human toxicity, cancer (where scores for hydrochar from organic fraction of MSW decrease by a factor of 3), freshwater ecotoxicity (where scores decrease by ca. 2 orders of magnitude across all waste streams, apart from digestate), and freshwater eutrophication (where scores decrease by ca. 1 order of magnitude across all waste streams) (Table S41).

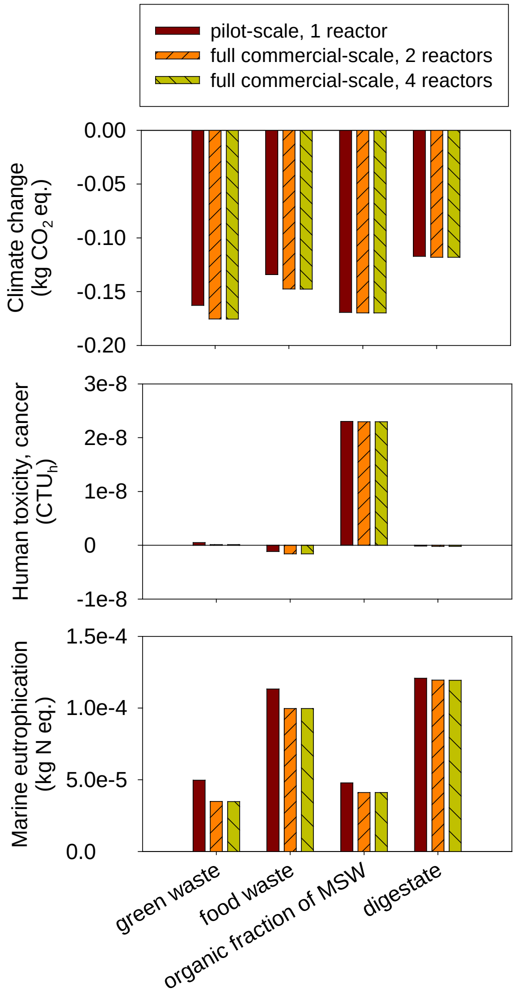

Fig. 2. Characterized impact scores in category-specific for three impact categories units including long-term emissions for each wet biomass waste stream treated hydrothermally at pilot- and full commercial-scale (scenarios 1-12 in Table 1).. Absolute uncertainties are too large to be shown, but statistical comparison taking into account correlation between uncertainties revealed significant differences between waste streams and plant scales (see SI†, Section S6).

3.3. Substituted waste management system and collection of biowaste influence performance

To explain the aforementioned trends and ultimately to identify improvement potentials for the HTC technology a process contribution analysis was conducted, i.e. identifying the processes with the largest environmental burden (Fig. 3). It shows that avoided generation of heat (i.e. heat that does not have to be generated from hard coal) is seen as important contributor as it avoids impacts stemming from combustion of hard coal briquettes, consistent with findings of Berge et al.[23] However, it also reveals two unusual trends: (i) positive contribution to total impacts from avoided waste management system, depending on the impact category; and (ii) high scores stemming from separate biowaste collection, depending on the biowaste type.

Avoided composting contributes to negative impact scores mainly due to avoided emissions of biogenic CH_4 and N_2O (for climate change), NO_x (for photochemical ozone formation and marine eutrophication), and NH_3 (for acidification and eutrophication). Contrarily, inclusion of credits for avoided production of inorganic (NPK) fertilizer in biowaste composting induces positive contributions when this process is avoided. When biowaste is carbonized hydrothermally these fertilizers will be produced using conventional techniques like the energy-intensive Haber-Bosch process for fixation of N from air (as system expansion is prioritized over allocation when handling multifunctional processes in our LCA study, consistent with the ISO standard and ILCD recommendations). For digestate, the conventional waste treatment option is incineration with energy recovery and the positive contribution from avoided incineration that is observed in some impact categories is due to system expansion performed to credit for the generated heat and electricity at the waste incinerator. This explains relatively poor performance of hydrochar produced from digestate when compared to hydrochars produced from other waste streams.

Processes of collection and transportation of waste are often omitted from system boundaries in LCA studies on waste management systems (they were omitted in 37% of all 200 published studies until 2014).[37,38] While impacts stemming from transportation of waste are usually not important contributor to total impacts for various waste treatment processes, our results show the contribution of biowaste collection to total impact scores for food waste is large (up to 50 % of total positive impact). This is because of the significant large transportation work required per unit of heat output from the stove, particularly when biowaste is very wet (e.g. food waste with 84% water content at collection point) and transportation distance is longer. Transportation work is also important for digestate despite its smaller water content as compared to the food waste because transportation distance is longer.. If transportation distances were the same for all biowaste streams (and equal to 7 km which is the distance for green waste, which had the shortest transportation distance across all four biowaste streams), the performance of food waste improves and food waste is seen best or second best in 1 and

6 impact categories, respectively (SI†, Section S6, Tables S43 and S44). In contrast, ranking of hydrochars made from organic fraction of MSW does not change that much (they are each seen the worst in 6 impact categories, compared to 5 and 7 categories in the baseline for organic fraction of MSW and digestate, respectively). This shows that: (i) the contribution of transportation work to total environmental impact can be large as it is influenced by both water content of the biowaste and transportation distance, and (ii) this important contribution from transportation work can influence ranking of hydrochar systems in terms or environmental impacts when biowaste is very wet (> 80% water content).

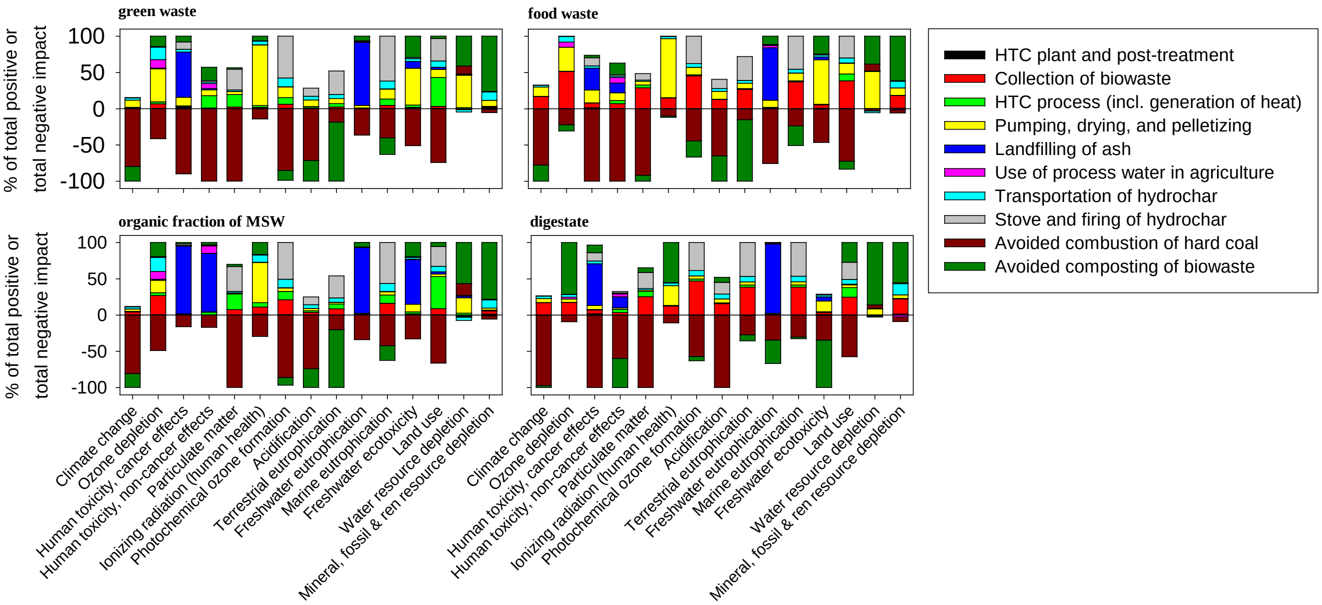

Fig. 3. Contribution of life cycle processes to total impacts from hydrothermal carbonization of four wet biomass waste streams at pilot scale. The scores for each impact category are scaled to 100% for categories with a net positive impact and to or -100% for impact categories where the net impact score is negative (i.e. avoided impacts are larger than induced impact). Long-term emissions are included. Note that the “HTC plant and post-treatment” category includes material input for construction of HTC installation together with end of life treatment processes, while HTC process includes generation of heat for running of the HTC process and emissions from the reactor.

3.4. The role of biowaste type and properties

Biowaste composition influences environmental performance in three ways: (i) through direct emissions from disposal of post-combustion ash; (ii) through direct emissions from spreading of process water on the soil; and (iii) through direct emissions from hydrochar combustion in the stove. In addition, water content of the feedstock that is pumped into the reactor influences environmental performance through indirect emissions stemming from processes associated with generation of electricity for pumping.

Differences in content of toxic metallic elements in the post-treatment and post-combustion ashes explain one and three orders of magnitude differences in impact scores for the toxic impacts on human health and ecotoxic impacts on freshwater ecosystems, respectively. Indeed, across all biowaste streams, the largest impact scores are reported for the system where hydrochar is produced from the organic fraction of MSW, mainly due to landfilling of contaminated ash as the organic fraction of MSW is contaminated with toxic metallic elements (like toxic cadmium and arsenic) originating from other MSW fractions. Concentrations of metallic elements in the process water from HTC of organic fraction of MSW are also higher (by ca. one order of magnitude, see SI†, Section S3), which further contributes to higher toxic impact scores for this type of hydrochar system. Berge et al.[23] also showed that HTC process water emissions are important contributors to impact scores for the toxicity-related impact categories, but they did not include emissions from hydrochar solids. Our results show that short- and long-term impacts from disposal of ashes are even more important than process water emissions, irrespective of the biowaste type and fraction of metals associated with hydrochar solid phase. They also show that the use of process water as fertilizer has the potential to increase human health impacts (non-cancer) due primarily to its contents of metals like zinc which are spread on the soil together with the process water, and to increase freshwater eutrophication impacts from phosphate emissions (both modelled as direct emission to soil). Although the use of process water as fertilizer in a citrus plantation allows for avoiding impacts stemming from fertilizer production, most notably impacts associated with resource depletion (for P), the extent of this reduction is very small compared to the contribution from impacts stemming from the need to produce fertilizers using conventional processes as a consequence of not producing compost.

Potential toxic impacts arising from emissions associated with combustion of hydrochar produced from organic fraction of MSW in a domestic stove are also up to one order of magnitude higher when compared to other biowaste streams, but this is not apparent in Fig. 3 because contribution from disposal of ash and direct emissions from process water is much larger. In addition, higher content of N and S in the hydrochar derived from organic fraction of MSW explains why acidification and eutrophication impacts in terrestrial ecosystems are higher as compared to hydrochar derived from other waste streams. Finally, firing of cleaned hydrochar in domestic stove contributes to the impact categories related to particulate matter and NO_x emissions. Conditions in the stove influence NO_x emissions, but they also depend on the content of organic-related nitrogen in the hydrochar, which is high for hydrochars derived from plant material like garden pruning in the green waste.

For many impact categories, the contribution from generation of electricity for running of the HTC plant and post-processing of the resulting hydrochar has also an important contribution to total environmental impact (Fig. 3). This contribution depends largely on the water content of the feedstock. While water content of the biowaste influences performance through its control of impacts stemming from biowaste collection, water content of the feedstock largely determines impacts through its control of the electricity used for pumping of the feedstock into the reactor. This demand is the highest for the food waste and the green waste feedstocks, which are very wet (>80% water content) (see SI†, Section S3).

3.5. Environmental performance at full-commercial scale

Comparison of impact scores between our LCA study and the study of Berge et al.[23] to investigate the effect of upscaling from lab-scale to pilot- or full commercial-scale are not possible because different processes were included in system boundaries. However, the environmental performance of HTC is expected to improve with upscaling. Indeed, Tables S42 and S43 (SI†, Section S6) show that with few exceptions impact scores for hydrochars produced from biowaste decrease with increasing plant scale due to reduced demand for heat and electricity. The differences are statistically significant in all impact categories apart from human toxicity (non-cancer) and water depletion. However, increasing plant configuration from two to four reactors does not improve environmental performance, with minor decreases in the impact scores with increasing capacity. This is because material input for construction of HTC installation is not an important contributor to total impacts and the main benefits from upscaling in the HTC plant are primarily due to a more efficient use of energy rather than sole size effects of the HTC installations. This is in contrast to technologies where material input for construction is an important driver of environmental impact, like wind power technology.[20] Thus, the largest improvement potentials lie in optimizing the use of heat and electricity use as plant scale increases, rather than optimizing material in the HTC installations. Our finding about small contribution from material inputs to total impacts also suggests that our conclusions are not affected by the upscaling factors used to estimate material needs from pilot- to the full commercial scale. When the technology matures, learning and experience with the technology over time might further contribute to improved environmental performance. Note, that for other types of HTC installations changes in dimension or types of reactors might be considered rather than adding more reactors of similar capacity and the same type, which might influence quality of the resulting hydrochar due to differences in process design. The consequences of such a change in hydrochar quality (in terms of change in HHV) would be linear response in environmental impact scores when HHV changes for the functional unit that is based on 1 MJ of heat output.

3.6. Is HTC an environmentally sound approach to treatment of biowaste?

HTC in Spain with hydrochar replacing hard coal briquettes is associated with -0.54 kg CO_2 eq per 1 kg of wet green waste treated. This result is in the range of anaerobic digestion with biogas recovery and incineration with energy recovery (i.e., -0.19 kg CO_2 eq per 1kg for anaerobic digestion and -0.093 per kg CO_2 eq per 1 kg for incineration, respectively) and is smaller compared other, more polluting treatment options (0.035 kg CO_2 eq per 1 kg for landfilling; 0.15 kg CO_2 eq per 1kg for composting).[33] Thus, treatment of 1 kg of wet green waste using HTC in Spain brings ca. three and six times more benefits even when substituting with the best (from the climate change perspective) alternative treatment options. Berge et al.[23] already showed that the type of fuel replaced by the hydrochar influences the environment performance. Here, we corroborate their study by showing that the substituted waste management system and composition of the electricity mix are also important for the environmental performance of HTC. Indeed, hydrochar derived from green waste (with HTC replacing incineration with energy recovery) performs worse when it is produced and used in Germany, with impact scores being significantly higher in 7 impact categories (see Tables S44 and S45 of the SI†, Section 6), including climate change. By contrast, for the digestate (for which incineration with energy recovery is the regular alternative in both Spain and Germany) the differences between Spain and Germany are in 14 out of 15 cases not statistically significant. The reader should note, that our findings about worse environmental performance of hydrochar produced from green waste in Germany apply to the current composition of the German electricity mix. If the future German mix includes cleaner energy sources (e.g. increasing the share of wind power to the grid), recovery of energy at the incinerator when biowaste is incinerated will substitute cleaner energy, in which case HTC will become more competitive when hydrochar replaces fossil-based fuels.

In summary, our LCA results point to the conclusion that HTC of biowaste with energy recovery when hydrochar is used as solid fuel may be an attractive treatment options for biowaste, depending on geographic location and substituted waste management system, with potential for further optimization. They corroborate earlier studies concluding that generalization of LCA results across different geographic locations should be done with caution.[37,38]

3.7. Recommendations to technology developers and data gaps

Our findings highlight the need for considering water content of the biowaste and that of the feedstock when optimizing environmental performance of HTC plants. Designers might influence transportation work and pumping efficiency of the feedstock; water content in the collection should be kept low to reduce transport work (below 50% for distances up to 20-30 km), while in the processing in the HTC it should be kept at a level which gives the best pumping efficiency of the feedstock. Trials with four types of biomass at pilot-scale show that the best pumping efficiency in terms of electricity used is achieved for feedstocks with water content of ca. 60% (see SI†, Section S3).

Composition of the biowaste to a large extent determines the environmental performance of HTC. This finding is generally in agreement with that of Berge et al.,[23] although inclusion of more processes within our system boundaries points to different direction with regard to main drivers of impacts. Namely, focus should be put on finding ways of utilizing ash separated from the hydrochar as short- and long-term emissions from ash disposal determine the magnitude of impact scores in toxicity-related impact categories. To help minimize these impacts, technology developers may consider employing more efficient cleaning in the post-treatment phase of the HTC process, like chemical cleaning using acid or alkali-acid leaching procedures.[39,40]

Detailed data about composition and fate of gases emitted from the HTC reactor(s) should be determined by technology developers as these potentially may contain compounds that are toxic and thus not negligible for the overall environmental performance of the HTC. Berge et al.[23] measured that various gases form during hydrothermal carbonization, including NMVOC and furans. In the current configuration of the HTC plant, these gases are directed into the boiler, but if their combustion in the boiler is incomplete, toxic impacts on human health will be underestimated.

Caution should be used when applying of process water in agriculture as it contains potentially toxic metals. We note, however, that there is uncertainty about both the composition of the process water, and the potential benefits from apparent increases in crop yield. We thus recommend technology developers to measure the composition of process water with respect to content of potentially beneficial (for crop growth) organic compounds. Berge et al.[1] measured various organic compounds in process water, but their inclusion in the current study was not possible due to incomplete knowledge about the compounds emitted and (likely) missing characterization factors. We do not expect that this limitation will influence impact scores to the extent that would change our conclusions because characterization factors for organic substances are usually much lower as compared to metals.[41–43]

If hydrochar is used as solid fuel, technology developers should focus on providing robust data emissions of potentially toxic metallic elements from combustion in the stove for various combustion parameters. Here, we adapted existing ecoinvent processes to model emissions resulting from combustion of the hydrochar assuming that transfer coefficients are the same (while correcting for differences in composition between biowaste types). The uncertainty analysis explored this data gap, resulting in expected spread to be within one to two orders of magnitude around actual values. This uncertainty is smaller as compared to the uncertainty in freshwater and human toxicity characterization factors, which is about three orders of magnitude.[44] Consideration of uncertainty in characterization factors was outside the goal of the study, but it is not expected to influence our conclusions about major drivers of environmental impacts, although it might change ranking of the four waste streams for freshwater ecotoxicity and human health impact categories.[41]

Finally, from environmental performance perspective, higher inputs of materials for HTC installations can be justified if they allow for increasing optimization of the plant in terms of heat and electricity use (e.g. during pumping, drying, and pelletizing) and thereby increasing environmental benefits associated with hydrothermal treatment. Our study for two- and four-reactor full-scale configurations displayed this, whereby larger material inputs (per unit of biowaste treated) do not translate into higher environmental impacts due small contribution of material to total impact.

Supporting Information. Expected changes introduced by upscaling on the environmental performance of HTC; life cycle impact assessment methods and normalization factors; parameters and data underlying LCA model; unit processes and LCI results; uncertainty factors and squared geometric standard deviations; and additional LCIA results.

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TOC

Title: Environmental performance of hydrothermal carbonization of four wet biomass waste streams at industry-relevant scales

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Synopsis: Life cycle assessment to support environmentally conscious design and installation of future HTC plants