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Additional Information

1 **Environmental performance of hydrothermal carbonization of four wet** 2 **biomass waste streams at industry-relevant scales**

3 Mikołaj Owsianiak^{a*}, Morten W. Ryberg^a, Michael Renz^b, Martin Hitzl^c, Michael Z. Hauschild^a

4
5 ^a Division for Quantitative Sustainability Assessment, Department of Management Engineering,
6 Technical University of Denmark, Produktionstorvet, Building 424, DK-2800 Kgs. Lyngby, Denmark

7 ^b Instituto de Tecnología Química (UPV-CSIC), Universitat Politècnica de València- Consejo Superior
8 de Investigaciones Científicas, Avenida de los Naranjos s/n,46022 Valencia, Spain

9 ^c Ingelia, S.L., C/Jaime Roig 19, 46010 Valencia, Spain

10 * miow@dtu.dk

11 +45 4525 4805

12 13 **Abstract**

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

28 **Keywords**

29 bioenergy, biowaste, hydrochar, life cycle assessment, upscaling

30 **1. Introduction**

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

39 During HTC, biomass is dehydrated in the presence of water by applying temperature (around
40 180-250 °C) and pressure (around 10-20 bars).^{1,8} The main products of HTC are the carbonaceous
41 material hydrochar, process water containing various inorganic and organic compounds, and non-
42 condensable gases.^{9,10} Hydrochar has properties that make it a good candidate for use as solid fuel, soil
43 conditioner with carbon storage value, or a material for construction of battery electrodes.¹¹⁻¹⁸ HTC
44 plants are based on either vertical (e.g. AVA-CO2, TerraNova Energy GmbH) or horizontal reactors
45 (e.g. Artec Biotechnologie GmbH, Grenol GmbH) in various configurations. The Spanish small-
46 medium enterprise (SME) Ingelia S.L., has erected one of the first pilot-scale HTC plants, that employs
47 one cylindrical vertical reactor operating continuously.⁹ Wet biomass is fed from the bottom, the
48 resulting hydrochar/water slurry is removed (also from the bottom) while the gases are collected from
49 the top. To increase capacity, the SME plans to add a second reactor, and furthermore, more two- and
50 four-reactor plants (with larger reactors relative to the pilot-scale reactor) will be installed in a near
51 future in other countries. Other HTC technology developers also allow for upscaling of their plants and
52 offer modular design of HTC installations (e.g. AVA-CO2).

53 Environmental performance of HTC is expected to change when upscaling to the full
54 commercial-scale is done.¹⁹⁻²¹ Table S1 of the SI†, Section S1, shows the potential implications of
55 upscaling on environmental performance of HTC of biowaste. For example, higher input of steel,
56 metals and crude oil per unit of plant is expected to cause linear increase of the impacts on climate
57 change, resource depletion, and various toxicity- and non-toxicity related impact categories due to the
58 need for manufacturing of additional reactors and plant equipment. Antagonistically, non-linear
59 capacity increase as dimensions or reactors change and plant grows (resulting in higher hydrochar

60 output per unit of plant) is expected to decrease these impacts, depending on the contribution of the
61 plant materials to total life cycle impacts. Thus, an assessment of environmental performance of the
62 technology must also consider the effects of size and capacities of the plant. Further, the environmental
63 performance of HTC is expected to be influenced by the regular waste management system that HTC
64 replaces.²² Neither pilot- nor full commercial-scale performance of HTC considering these factors has,
65 to date, been assessed using LCA.

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

81 In this paper, we address these four limitations, and concurrently present life cycle inventory
82 (LCI) and life cycle impact assessment (LCIA) results of HTC of green waste (being garden
83 trimmings), food waste (represented by orange peels), organic fraction of municipal solid waste and
84 digestate at industry-relevant scales. All waste-streams are promising candidates for hydrothermal
85 carbonization at full commercial-scale as validated by a pilot-scale assessment. Primary data from
86 pilot-plant operations were used to model the foreground processes. Emissions of CO₂, CO, nitrous
87 oxides (NO_x), SO₂ and particulate matter (PM) from hydrochar combustion were based on
88 measurements, while emissions of metals were taken from genericecoinvent process for incineration of
89 biowaste while correcting for differences in composition and properties between hydrochar and

90 biowaste. To illustrate the potential of the technology, environmental performance at pilot-scale with
91 one reactor was compared to that at full commercial scale with two or four reactors. Full-scale plants
92 differ from the pilot-scale mainly with regard to plant capacity (increasing capacity with increasing
93 scale, resulting from increasing the number and dimensions of reactors) and energy and material inputs
94 (decreasing inputs per treated quantity with increasing scale).

95

96 **2. Methods**

97 **2.1. Wet biomass waste streams**

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

118

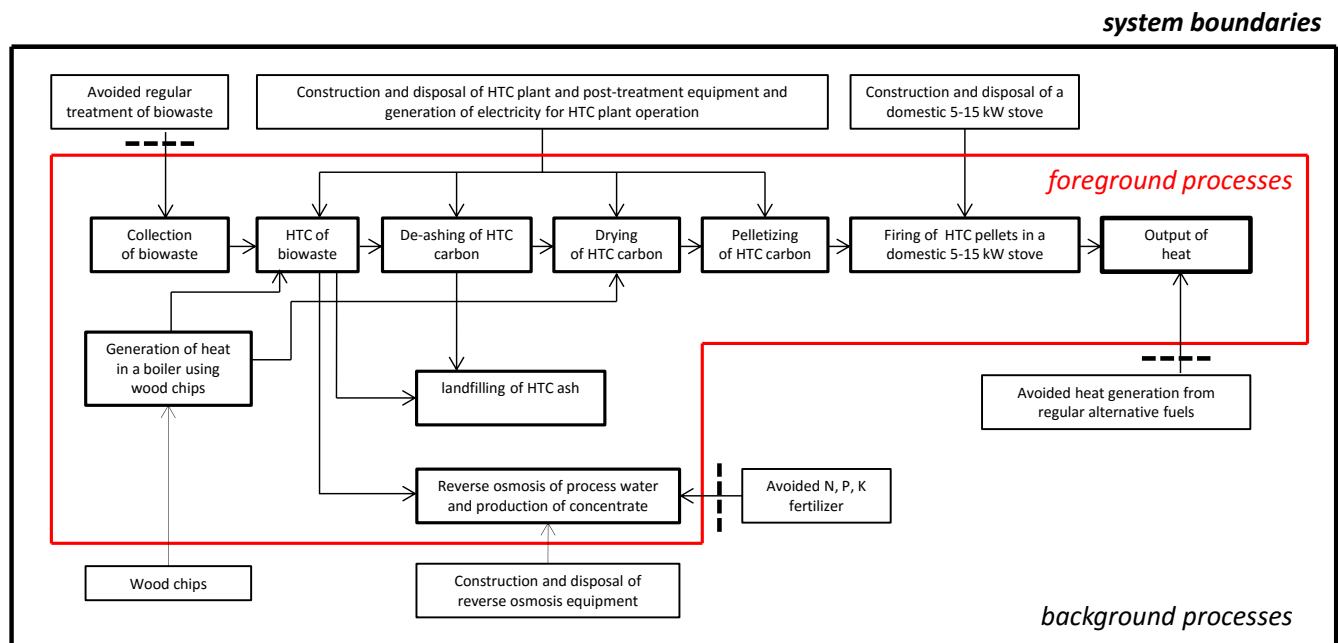
119 **2.2. Life cycle assessment**

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

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

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

138



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

142

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

152

153 Table 1. Overview of the compared scenarios for hydrothermal carbonization (HTC) wet biomass
 154 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)

155 ^a ES: Spain; UK: the United Kingdom; DE: Germany

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

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

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

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

169

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

184

185 **Life cycle impact assessment.** The product systems were modeled in SimaPro, version 8.0.4.30 (PRé
186 Consultants bv, the Netherlands). Environmental impact scores were calculated using the ILCD’s
187 recommended practice characterization factors at midpoint (ILCD 2011 Midpoint+, version 1.05), as
188 implemented in SimaPro.³² This recommended practice has been identified by assessing a total of 156
189 different characterization models belonging to 12 different LCIA methodologies.³² All ILCD impact
190 categories were considered, apart from ionizing radiation impacts on ecosystems which considered not

191 sufficiently representative for this type of impact. Ranking of biowaste streams may be sensitive to the
192 inclusion/exclusion of long-term emissions (that is, emissions occurring after 100 years) which may
193 determine the magnitude of eutrophication- and toxicity-related impact scores. Since the long-term
194 emissions have larger uncertainties than short-term emissions, it is of interest to see how their inclusion
195 affects the conclusions. Thus, the impact scores were calculated with long-term emissions either
196 included (default settings) or excluded from the assessment. Normalization was done using the
197 European set of ILCD's normalization factors for reference year 2010, version 4.0, as implemented in
198 SimaPro. Synthesis of the LCIA methods and normalization factors are presented in SI†, Section S2.

199

200 **2.3. Data and model parameters**

201 Unit processes for the foreground system were constructed using model parameters based on
202 measurements performed at a pilot plant at Ingelia S.L. (Valencia, Spain). They are synthesized in the
203 SI†, Section S3. Background information of the plant itself is given elsewhere.⁹ We measured
204 parameters related to: (i) composition of the biowaste (i.e., content of water, ash, nitrogen (N), carbon
205 (C), sulfur (S)); (ii) HTC plant and post-treatment equipment (i.e., material inputs, plant utilization rate,
206 overall plant capacity, electricity and heat use for pumping of feedstock, electricity use for drying and
207 pelletizing, yield of raw hydrochar and yield of hydrochar pellets, amount of process water, amount of
208 gases); (iii) properties of hydrochar (i.e., content of water, ash, N, C, S, fluoride, chloride, and higher
209 heating value of hydrochar pellets); (iv) combustion of hydrochar pellets (i.e., emissions of CO₂, CO
210 particulate matter (PM); and (v) composition of the ash (phosphor (P), boron (B), and 19 metals and
211 metalloids), composition of process water (N, P, B, and 19 metals and metalloids); and composition of
212 gases (CO₂, CO, H₂). Emissions of PM, CO₂, CO, nitrogen oxides (NO_x), and SO₂ from hydrochar
213 combustion in the stove are based on measurements performed during experiments using a pilot-scale
214 (180 kW) grate combustion unit. Emissions of metallic elements to air were calculated using transfer
215 coefficients for emissions to air from the ecoinvent process for incineration of biowaste, corrected for
216 differences in composition and moisture between the hydrochar and the biowaste in the ecoinvent
217 process. Life times of HTC plant, post-treatment equipment, reverse osmosis membrane, and buildings,
218 and thermal efficiency of the boiler, were assumed using values based on reasonable expectations.
219 Transportation distances between the plant, retail, and final user were taken from Google maps,
220 whereas location of the final user (in the UK) is unknown and had to be assumed.

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

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

241

242 **2.4. Uncertainty analysis**

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

251 foreground processes. Uncertainties in the background processes were based on geometric standard
252 deviations already assigned to flows in the ecoinvent processes.

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

259

260 **3. Results and discussion**

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

266

267 **3.1. Life cycle inventories**

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

274

275 **3.2. Overview of life cycle impact assessment results**

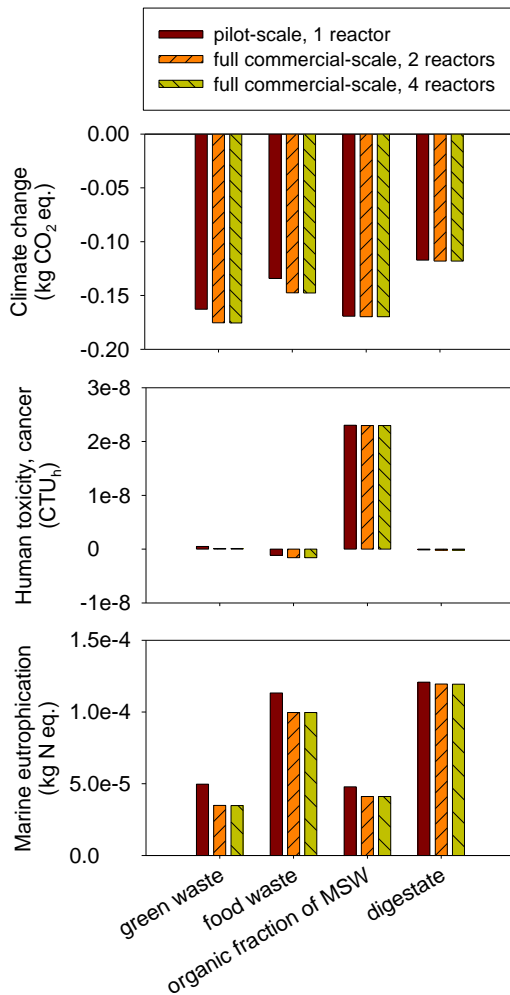
276 Figure 2 shows characterized life cycle impact assessment (LCIA) results for the baseline scenario and
277 the scenario showing the influence of plant scale for four selected impact categories. They represent
278 typical impact profiles observed for the four wet biomass waste streams. The results scores for other
279 scenarios across all 15 ILCD's impact categories are presented in SI†, Section S6 (Tables S39-S47).
280 The LCIA results show four main trends. First, the impact scores are negative for 6 (green waste, food

281 waste, and digestate) and 5 (organic fraction of MSW) impact categories (Table S39 and S40). s Third,
282 green waste is seen as the best or second best in 2 and 10 impact categories, respectively, and
283 statistically significant differences in impact scores between green waste and food waste, organic
284 fraction of MSW, and digestate occur in 14, 12, and 6 impact categories, respectively. Fourth, digestate
285 is seen the worst in 7 impact categories although in 10 impact categories the difference between
286 digestate and food waste is not statistically significant (Table S39 and S40).

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

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

302



303

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

309

310

311 3.3. Substituted waste management system and collection of biowaste influence performance

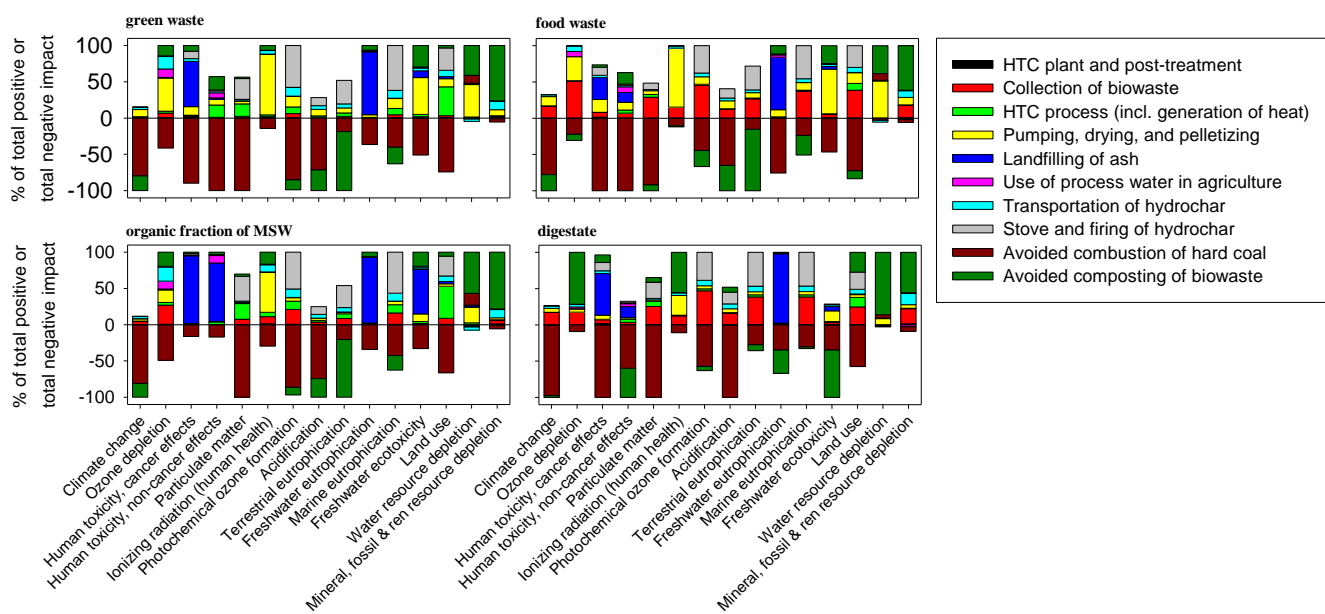
312 To explain the aforementioned trends and ultimately to identify improvement potentials for the HTC
 313 technology a process contribution analysis was conducted, i.e. identifying the processes with the largest
 314 environmental burden (Fig. 3). It shows that avoided generation of heat (i.e. heat that does not have to

315 be generated from hard coal) is seen as important contributor as it avoids impacts stemming from
316 combustion of hard coal briquettes, consistent with findings of Berge et al.²³ However, it also reveals
317 two unusual trends: (i) positive contribution to total impacts from avoided waste management system,
318 depending on the impact category; and (ii) high scores stemming from separate biowaste collection,
319 depending on the biowaste type.

320 Avoided composting contributes to negative impact scores mainly due to avoided emissions of
321 biogenic CH₄ and N₂O (for climate change), NO_x (for photochemical ozone formation and marine
322 eutrophication), and NH₃ (for acidification and eutrophication). Contrarily, inclusion of credits for
323 avoided production of inorganic (NPK) fertilizer in biowaste composting induces positive contributions
324 when this process is avoided. When biowaste is carbonized hydrothermally these fertilizers will be
325 produced using conventional techniques like the energy-intensive Haber-Bosch process for fixation of
326 N from air (as system expansion is prioritized over allocation when handling multifunctional processes
327 in our LCA study, consistent with the ISO standard and ILCD recommendations). For digestate, the
328 conventional waste treatment option is incineration with energy recovery and the positive contribution
329 from avoided incineration that is observed in some impact categories is due to system expansion
330 performed to credit for the generated heat and electricity at the waste incinerator. This explains
331 relatively poor performance of hydrochar produced from digestate when compared to hydrochars
332 produced from other waste streams.

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

345 6 impact categories, respectively (SI†, Section S6, Tables S43 and S44). In contrast, ranking of
 346 hydrochars made from organic fraction of MSW does not change that much (they are each seen the
 347 worst in 6 impact categories, compared to 5 and 7 categories in the baseline for organic fraction of
 348 MSW and digestate, respectively). This shows that: (i) the contribution of transportation work to total
 349 environmental impact can be large as it is influenced by both water content of the biowaste and
 350 transportation distance, and (ii) this important contribution from transportation work can influence
 351 ranking of hydrochar systems in terms of environmental impacts when biowaste is very wet (> 80%
 352 water content).



353
 354 Fig. 3. Contribution of life cycle processes to total impacts from hydrothermal carbonization of
 355 four wet biomass waste streams at pilot scale. The scores for each impact category are scaled to 100%
 356 for categories with a net positive impact and to or -100% for impact categories where the net impact
 357 score is negative (i.e. avoided impacts are larger than induced impact). Long-term emissions are
 358 included. Note that the “HTC plant and post-treatment” category includes material input for
 359 construction of HTC installation together with end of life treatment processes, while HTC process
 360 includes generation of heat for running of the HTC process and emissions from the reactor.

361 3.4. The role of biowaste type and properties

362 Biowaste composition influences environmental performance in three ways: (i) through direct
 363 emissions from disposal of post-combustion ash; (ii) through direct emissions from spreading of

364 process water on the soil; and (iii) through direct emissions from hydrochar combustion in the stove. In
365 addition, water content of the feedstock that is pumped into the reactor influences environmental
366 performance through indirect emissions stemming from processes associated with generation of
367 electricity for pumping.

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

389 Potential toxic impacts arising from emissions associated with combustion of hydrochar produced
390 from organic fraction of MSW in a domestic stove are also up to one order of magnitude higher when
391 compared to other biowaste streams, but this is not apparent in Fig. 3 because contribution from
392 disposal of ash and direct emissions from process water is much larger. In addition, higher content of N
393 and S in the hydrochar derived from organic fraction of MSW explains why acidification and

394 eutrophication impacts in terrestrial ecosystems are higher as compared to hydrochar derived from
395 other waste streams. Finally, firing of cleaned hydrochar in domestic stove contributes to the impact
396 categories related to particulate matter and NO_x emissions. Conditions in the stove influence NO_x
397 emissions, but they also depend on the content of organic-related nitrogen in the hydrochar, which is
398 high for hydrochars derived from plant material like garden pruning in the green waste.

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

407

408 **3.5. Environmental performance at full-commercial scale**

409 Comparison of impact scores between our LCA study and the study of Berge et al.²³ to investigate the
410 effect of upscaling from lab-scale to pilot- or full commercial-scale are not possible because different
411 processes were included in system boundaries. However, the environmental performance of HTC is
412 expected to improve with upscaling. Indeed, Tables S42 and S43 (SI†, Section S6) show that with few
413 exceptions impact scores for hydrochars produced from biowaste decrease with increasing plant scale
414 due to reduced demand for heat and electricity. The differences are statistically significant in all impact
415 categories apart from human toxicity (non-cancer) and water depletion. However, increasing plant
416 configuration from two to four reactors does not improve environmental performance, with minor
417 decreases in the impact scores with increasing capacity. This is because material input for construction
418 of HTC installation is not an important contributor to total impacts and the main benefits from
419 upscaling in the HTC plant are primarily due to a more efficient use of energy rather than sole size
420 effects of the HTC installations. This is in contrast to technologies where material input for
421 construction is an important driver of environmental impact, like wind power technology.²⁰ Thus, the
422 largest improvement potentials lie in optimizing the use of heat and electricity use as plant scale
423 increases, rather than optimizing material in the HTC installations. Our finding about small

424 contribution from material inputs to total impacts also suggests that our conclusions are not affected by
425 the upscaling factors used to estimate material needs from pilot- to the full commercial scale. When the
426 technology matures, learning and experience with the technology over time might further contribute to
427 improved environmental performance. Note, that for other types of HTC installations changes in
428 dimension or types of reactors might be considered rather than adding more reactors of similar capacity
429 and the same type, which might influence quality of the resulting hydrochar due to differences in
430 process design. The consequences of such a change in hydrochar quality (in terms of change in HHV)
431 would be linear response in environmental impact scores when HHV changes for the functional unit
432 that is based on 1 MJ of heat output.

433

434 **3.6. Is HTC an environmentally sound approach to treatment of biowaste?**

435 HTC in Spain with hydrochar replacing hard coal briquettes is associated with -0.54 kg CO₂ eq per 1
436 kg of wet green waste treated. This result is in the range of anaerobic digestion with biogas recovery
437 and incineration with energy recovery (i.e., -0.19 kg CO₂ eq per 1kg for anaerobic digestion and -0.093
438 per kg CO₂ eq per 1 kg for incineration, respectively) and is smaller compared other, more polluting
439 treatment options (0.035 kg CO₂ eq per 1 kg for landfilling; 0.15 kg CO₂ eq per 1kg for composting).³³
440 Thus, treatment of 1 kg of wet green waste using HTC in Spain brings ca. three and six times more
441 benefits even when substituting with the best (from the climate change perspective) alternative
442 treatment options. Berge et al.²³ already showed that the type of fuel replaced by the hydrochar
443 influences the environment performance. Here, we corroborate their study by showing that the
444 substituted waste management system and composition of the electricity mix are also important for the
445 environmental performance of HTC. Indeed, hydrochar derived from green waste (with HTC replacing
446 incineration with energy recovery) performs worse when it is produced and used in Germany, with
447 impact scores being significantly higher in 7 impact categories (see Tables S44 and S45 of the SI†,
448 Section 6), including climate change. By contrast, for the digestate (for which incineration with energy
449 recovery is the regular alternative in both Spain and Germany) the differences between Spain and
450 Germany are in 14 out of 15 cases not statistically significant. The reader should note, that our findings
451 about worse environmental performance of hydrochar produced from green waste in Germany apply to
452 the current composition of the German electricity mix. If the future German mix includes cleaner
453 energy sources (e.g. increasing the share of wind power to the grid), recovery of energy at the

454 incinerator when biowaste is incinerated will substitute cleaner energy, in which case HTC will become
455 more competitive when hydrochar replaces fossil-based fuels.

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

461

462 **3.7. Recommendations to technology developers and data gaps**

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

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

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

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

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

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

510

511 **Supporting Information.** Expected changes introduced by upscaling on the environmental
512 performance of HTC; life cycle impact assessment methods and normalization factors; parameters and

513 data underlying LCA model; unit processes and LCI results; uncertainty factors and squared geometric
514 standard deviations; and additional LCIA results.

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643 **TOC**

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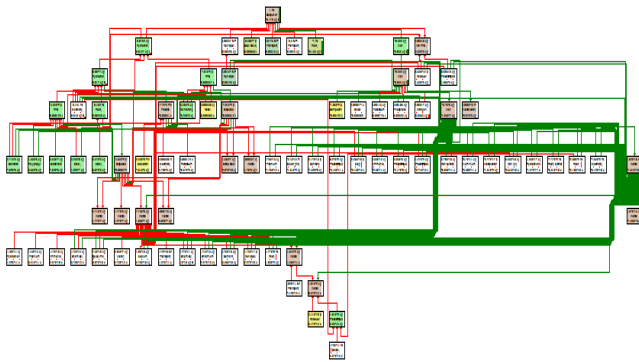
645 Title: Environmental performance of hydrothermal carbonization of four wet biomass waste streams at
646 industry-relevant scales

647 Authors: Mikołaj Owsianiak, Morten W. Ryberg, Michael Renz, Martin Hitzl, Michael Z. Hauschild

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

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