



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

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

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8

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16

17 **Abstract**

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

32 **Keywords**

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

## 34 **1. Introduction**

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

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

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

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

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

85 In this paper, we address these four limitations, and concurrently present life cycle inventory  
86 (LCI) and life cycle impact assessment (LCIA) results of HTC of green waste (being garden  
87 trimmings), food waste (represented by orange peels), organic fraction of municipal solid waste and  
88 digestate at industry-relevant scales. All waste-streams are promising candidates for hydrothermal  
89 carbonization at full commercial-scale as validated by a pilot-scale assessment. Primary data from  
90 pilot-plant operations were used to model the foreground processes. Emissions of CO<sub>2</sub>, CO, nitrous  
91 oxides (NO<sub>x</sub>), SO<sub>2</sub> and particulate matter (PM) from hydrochar combustion were based on  
92 measurements, while emissions of metals were taken from genericecoinvent process for incineration of  
93 biowaste while correcting for differences in composition and properties between hydrochar and

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

99

## 100 **2. Methods**

### 101 **2.1. Wet biomass waste streams**

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

122

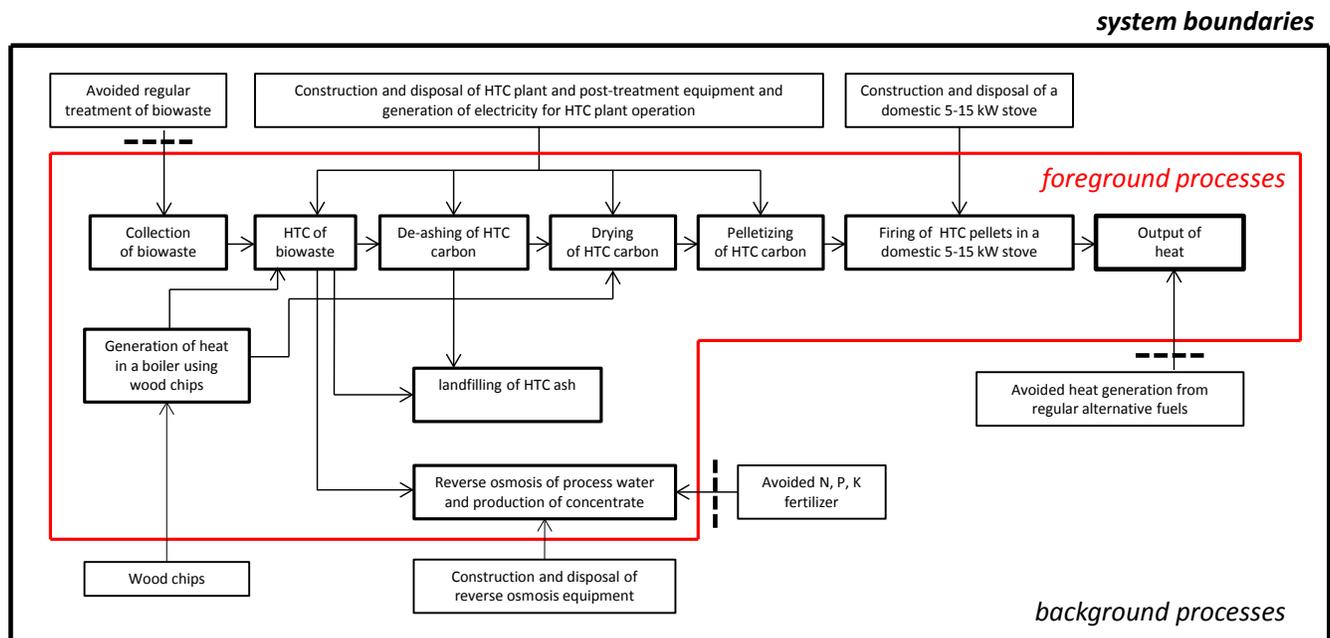
### 123 **2.2. Life cycle assessment**

124 The LCA was conducted in accordance with the requirements of the ISO standard and the guidelines of  
 125 the International Reference Life Cycle Data System (ILCD) handbook.<sup>29,30</sup>

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

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

142



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

146

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

156

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

# Scenario	Sensitivity parameter	Geographic location (production/use) <sup>a</sup>	Biowaste type <sup>c</sup>	Transportation distance of the biowaste to the plant (in km)	Plant scale <sup>b</sup>	Replaced waste management system (WMS) <sup>d</sup>
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)

159 <sup>a</sup> ES: Spain; UK: the United Kingdom; DE: Germany

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

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

164 <sup>c</sup> GW: green waste, FW: food waste, OFMSW: organic fraction of municipal solid waste, DG: digestate

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

173

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

188

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

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

203

### 204 **2.3. Data and model parameters**

205 Unit processes for the foreground system were constructed using model parameters based on  
206 measurements performed at a pilot plant at Ingelia S.L. (Valencia, Spain). They are synthesized in the  
207 SI†, Section S3. Background information of the plant itself is given elsewhere.<sup>9</sup> We measured  
208 parameters related to: (i) composition of the biowaste (i.e., content of water, ash, nitrogen (N), carbon  
209 (C), sulfur (S)); (ii) HTC plant and post-treatment equipment (i.e., material inputs, plant utilization rate,  
210 overall plant capacity, electricity and heat use for pumping of feedstock, electricity use for drying and  
211 pelletizing, yield of raw hydrochar and yield of hydrochar pellets, amount of process water, amount of  
212 gases); (iii) properties of hydrochar (i.e., content of water, ash, N, C, S, fluoride, chloride, and higher  
213 heating value of hydrochar pellets); (iv) combustion of hydrochar pellets (i.e., emissions of CO<sub>2</sub>, CO  
214 particulate matter (PM); and (v) composition of the ash (phosphor (P), boron (B), and 19 metals and  
215 metalloids), composition of process water (N, P, B, and 19 metals and metalloids); and composition of  
216 gases (CO<sub>2</sub>, CO, H<sub>2</sub>). Emissions of PM, CO<sub>2</sub>, CO, nitrogen oxides (NO<sub>x</sub>), and SO<sub>2</sub> from hydrochar  
217 combustion in the stove are based on measurements performed during experiments using a pilot-scale  
218 (180 kW) grate combustion unit. Emissions of metallic elements to air were calculated using transfer  
219 coefficients for emissions to air from theecoinvent process for incineration of biowaste, corrected for  
220 differences in composition and moisture between the hydrochar and the biowaste in the ecoinvent  
221 process. Life times of HTC plant, post-treatment equipment, reverse osmosis membrane, and buildings,  
222 and thermal efficiency of the boiler, were assumed using values based on reasonable expectations.  
223 Transportation distances between the plant, retail, and final user were taken from Google maps,  
224 whereas location of the final user (in the UK) is unknown and had to be assumed.

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

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

245

## 246 **2.4. Uncertainty analysis**

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

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

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

263

## 264 **3. Results and discussion**

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

270

### 271 **3.1. Life cycle inventories**

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

278

### 279 **3.2. Overview of life cycle impact assessment results**

280 Figure 2 shows characterized life cycle impact assessment (LCIA) results for the baseline scenario and  
281 the scenario showing the influence of plant scale for four selected impact categories. They represent  
282 typical impact profiles observed for the four wet biomass waste streams. The results scores for other  
283 scenarios across all 15 ILCD's impact categories are presented in SI†, Section S6 (Tables S39-S47).

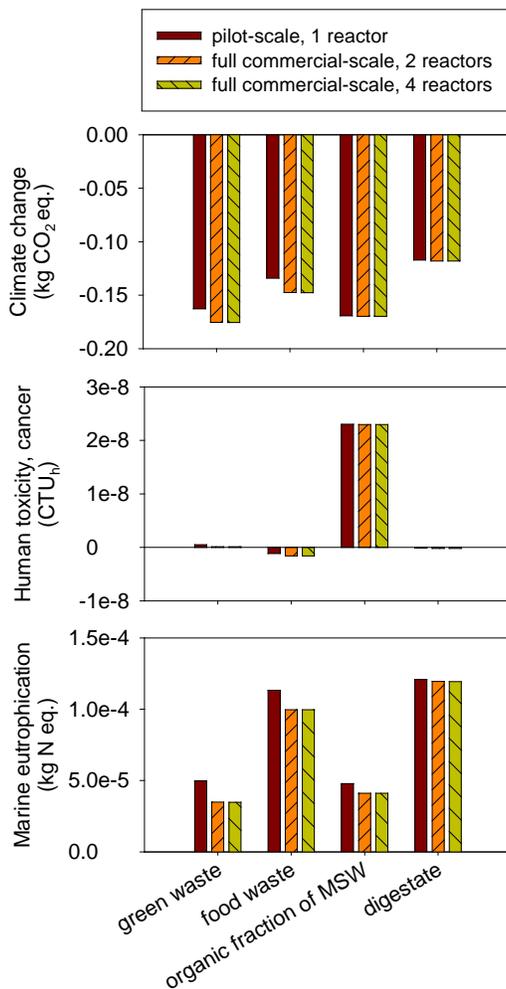
284 The LCIA results show four main trends. First, the impact scores are negative for 6 (green waste, food

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

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

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

306



307

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

313

314

### 315 3.3. Substituted waste management system and collection of biowaste influence performance

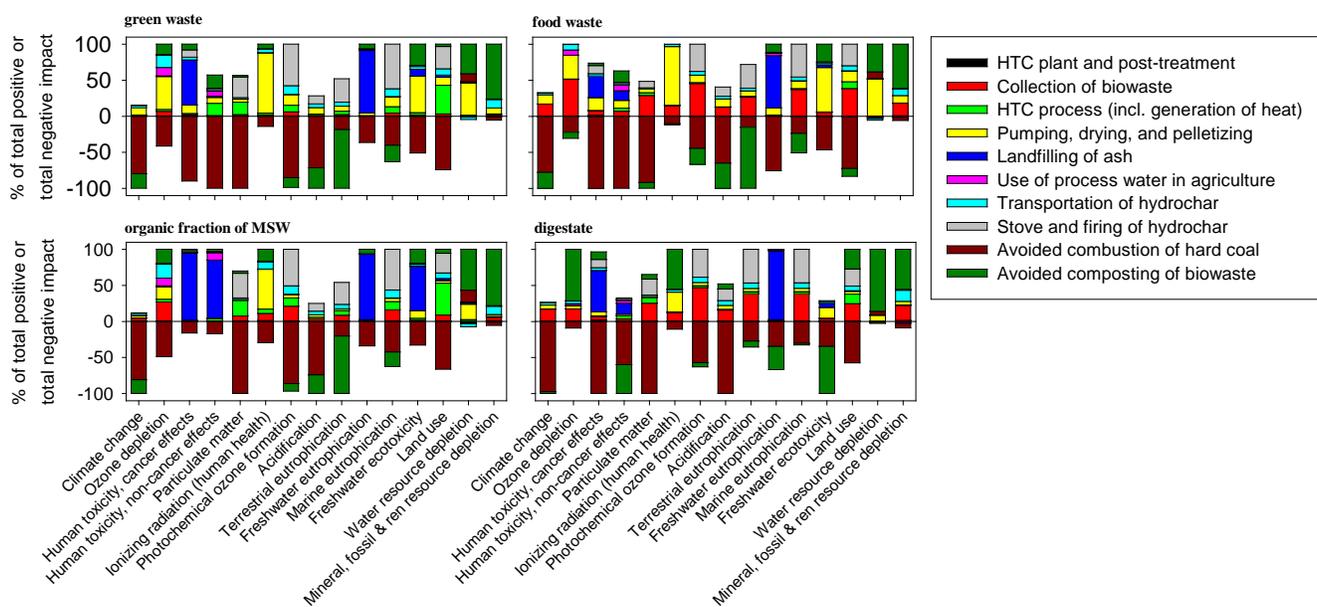
316 To explain the aforementioned trends and ultimately to identify improvement potentials for the HTC  
 317 technology a process contribution analysis was conducted, i.e. identifying the processes with the largest  
 318 environmental burden (Fig. 3). It shows that avoided generation of heat (i.e.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

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

324       Avoided composting contributes to negative impact scores mainly due to avoided emissions of  
325 biogenic CH<sub>4</sub> and N<sub>2</sub>O (for climate change), NO<sub>x</sub> (for photochemical ozone formation and marine  
326 eutrophication), and NH<sub>3</sub> (for acidification and eutrophication). Contrarily, inclusion of credits for  
327 avoided production of inorganic (NPK) fertilizer in biowaste composting induces positive contributions  
328 when this process is avoided. When biowaste is carbonized hydrothermally these fertilizers will be  
329 produced using conventional techniques like the energy-intensive Haber-Bosch process for fixation of  
330 N from air (as system expansion is prioritized over allocation when handling multifunctional processes  
331 in our LCA study, consistent with the ISO standard and ILCD recommendations). For digestate, the  
332 conventional waste treatment option is incineration with energy recovery and the positive contribution  
333 from avoided incineration that is observed in some impact categories is due to system expansion  
334 performed to credit for the generated heat and electricity at the waste incinerator. This explains  
335 relatively poor performance of hydrochar produced from digestate when compared to hydrochars  
336 produced from other waste streams.

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

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



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

### 365 3.4. The role of biowaste type and properties

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

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

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

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

398 eutrophication impacts in terrestrial ecosystems are higher as compared to hydrochar derived from  
399 other waste streams. Finally, firing of cleaned hydrochar in domestic stove contributes to the impact  
400 categories related to particulate matter and NO<sub>x</sub> emissions. Conditions in the stove influence NO<sub>x</sub>  
401 emissions, but they also depend on the content of organic-related nitrogen in the hydrochar, which is  
402 high for hydrochars derived from plant material like garden pruning in the green waste.

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

411

### 412 **3.5. Environmental performance at full-commercial scale**

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

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

437

### 438 **3.6. Is HTC an environmentally sound approach to treatment of biowaste?**

439 HTC in Spain with hydrochar replacing hard coal briquettes is associated with -0.54 kg CO<sub>2</sub> eq per 1  
440 kg of wet green waste treated. This result is in the range of anaerobic digestion with biogas recovery  
441 and incineration with energy recovery (i.e., -0.19 kg CO<sub>2</sub> eq per 1kg for anaerobic digestion and -0.093  
442 per kg CO<sub>2</sub> eq per 1 kg for incineration, respectively) and is smaller compared other, more polluting  
443 treatment options (0.035 kg CO<sub>2</sub> eq per 1 kg for landfilling; 0.15 kg CO<sub>2</sub> eq per 1kg for composting).<sup>33</sup>  
444 Thus, treatment of 1 kg of wet green waste using HTC in Spain brings ca. three and six times more  
445 benefits even when substituting with the best (from the climate change perspective) alternative  
446 treatment options. Berge et al.<sup>23</sup> already showed that the type of fuel replaced by the hydrochar  
447 influences the environment performance. Here, we corroborate their study by showing that the  
448 substituted waste management system and composition of the electricity mix are also important for the  
449 environmental performance of HTC. Indeed, hydrochar derived from green waste (with HTC replacing  
450 incineration with energy recovery) performs worse when it is produced and used in Germany, with  
451 impact scores being significantly higher in 7 impact categories (see Tables S44 and S45 of the SI†,  
452 Section 6), including climate change. By contrast, for the digestate (for which incineration with energy  
453 recovery is the regular alternative in both Spain and Germany) the differences between Spain and  
454 Germany are in 14 out of 15 cases not statistically significant. The reader should note, that our findings  
455 about worse environmental performance of hydrochar produced from green waste in Germany apply to  
456 the current composition of the German electricity mix. If the future German mix includes cleaner  
457 energy sources (e.g. increasing the share of wind power to the grid), recovery of energy at the

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

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

465

### 466 **3.7. Recommendations to technology developers and data gaps**

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

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

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

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

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

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

514

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

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

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523

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647 **TOC**

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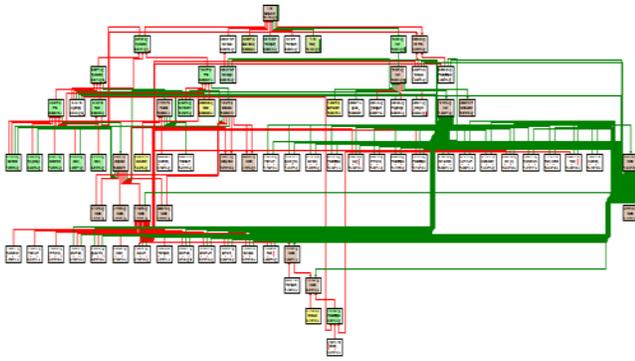
649 Title: Environmental performance of hydrothermal carbonization of four wet biomass waste streams at  
650 industry-relevant scales

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

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

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