

# Life cycle assessment (LCA) of end-of-life dairy products (EoL-DPs) valorization via anaerobic co-digestion with agro-industrial wastes for biogas production

Article

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1	Life Cycle Assessment (LCA) of End-of-Life Dairy Products (EoL-DPs) valorization via
2	anaerobic co-digestion with agro-industrial wastes for biogas production
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19 20	Declarations of interest: none

21	BACKGROUND: The aim of the present study was to assess the environmental impacts
22	of End-of-Life Dairy Products (EoL-DPs) management via their co-treatment with agro-
23	industrial wastes (AgW) in a centralized biogas facility located in Cyprus using a gate-
24	to-gate LCA approach. Two different scenarios were examined under the framework of
25	this project. In the first one, co-treatment of EoL-DPs with various AgW (in a 20/80,
26	w/w, ratio) was evaluated in a one-stage mesophilic anaerobic digestion (AD) process.
27	In the second scenario, the same amount of EoL-DPs were acidified before
28	methanogenesis with AgW in order to improve biogas production.
29	<b>RESULTS:</b> Prior acidification of EoL-DPs showed a better environmental performance
30	compared to the results obtained upon direct co-digestion in a mesophilic digester,
31	having a total impact of 52.44 Pt against 57.13 Pt respectively. Biogas production upon
32	acidification, and therefore energy yield, was higher reaching up to 22.88 $\mathrm{m^3}\mathrm{CH_4/ton}$
33	of feed (229.25 kWh/ton of feed), compared to 17.45 $m^3$ CH <sub>4</sub> /on of feed (174.85
34	kWh/ton of feed) for the case where no pretreatment was performed.
35	<b>CONCLUSIONS:</b> The acidification of EoL-DPs enhanced the environmental performance
36	of the process by reducing its impact by 8.2% (in Pt equivalents). The energy
37	consumption of the biogas plant mixing equipment was identified as the process
38	hotspot. However, further analysis of the environmental performance of the proposed
39	process is required by extending the system's boundaries towards a Cradle-to-Grave
40	approach.

- 42 Keywords: End-of-Life Dairy Products; Agro-industrial Wastes; Anaerobic Digestion; Life
- 43 Cycle Assessment; Bioenergy.

45 **INTRODUCTION** 

Nowadays, general scientific consensus believes that global warming is caused by the 46 emission of anthropogenic greenhouse gases (GHG), mainly derived from fossil fuel 47 48 combustion <sup>1</sup>. As a result, the demand for renewable energy is rising because of the 49 increasing social awareness of consequences related to non-renewable energy use, e.g. fossil fuel depletion, energy security, and climate change (CC). Renewable energy 50 51 production in the European Union is targeted to reach 20% and 27% of the total 52 energy production by 2020 and 2030 respectively <sup>2,3</sup>. This transition requires insight 53 into environmental alternatives of producing renewable energy, including CC, fossil 54 fuel depletion, and land use changes. Bioenergy is a renewable form of energy 55 produced from biomass, including energy crops, wood, microbial biomass as well as wastes from household, agriculture, cattle, forestry and industrial activities <sup>4</sup>. 56 57 Currently, there is a growing interest on the use of biomass for energy purposes in order to satisfy energy requirements all over Europe <sup>5</sup>. Since biomass accounts for 2/3 58 of the renewable energy produced in Europe, its valorization results in lower 59 60 dependency on fossil fuels for many European countries, depending on biomass local resources, in order to meet the renewable energy directive objectives <sup>6,3</sup>. 61 Biomass can be converted by anaerobic digestion (AD) into biogas, composed 62 63 of methane ( $CH_4$ ), carbon dioxide ( $CO_2$ ) and some trace gases (e.g., hydrogen). It is worth noting that in 2013 total biogas produced in Europe reached 14 billion m<sup>3</sup>, as in 64 natural gas equivalent, whereas the projection for 2020 is about 28 billion m<sup>3 7</sup>. Biogas 65 obtained can be exploited in situ to produce electricity or heat or preferably a 66 combination of both through cogeneration in a combined heat and power (CHP) unit. 67

On the other hand, it can be upgraded to the natural gas standards, in the form of biomethane, with a methane content up to 98%. Biomethane can be then forwarded to local natural gas distribution networks in order to be used for electricity and power generation. It can be also used for heating purposes either alone or blended with natural gas. Alternative scenarios include its application as a transportation fuel or a high-tech process energy and raw material for the chemical industry <sup>7,3</sup>.

74 Main substrates for AD include agricultural biomass, in the form of animal 75 manures and energy crops (e.g. maize, rye and grass silage), organic residues from 76 processing industries (e.g. glycerin, food waste, beet tails, slaughterhouse wastes etc.), 77 and other organic residues such as roadside grass, forest residues, sewage sludge etc.<sup>8</sup>. Those feedstocks are characterized by a methane content, in the produced 78 biogas, ranging between 51-72%<sup>9</sup>. Biogas has the potential to deliver more than 1/3 of 79 natural gas production in Europe and could reach about 15-25% of total bioenergy 80 81 produced by 2020, compared to 7% in 2007<sup>3</sup>. According to the European Biogas 82 Association, biogas plants in Europe increased by 3%, from 16,834 to 17,376, in 2015 83 and the total amount of electricity produced from biogas is approximately 63.3 TWh, corresponding to the annual consumption of 14.6 million European households <sup>10</sup>. 84 Germany has been in the lead, with 10,846 biogas plants, valorizing mainly agricultural 85 86 feedstocks (energy crops and agricultural residues), followed by Italy (1,555), France (717), Switzerland (638), Czech Republic (554) and UK (523) <sup>10,11</sup>. By the end of 2015, 87 88 fourteen biogas plants were operating in Cyprus, based on agricultural feedstocks. Their installed electrical capacity was approximately 10 MW<sub>el</sub>, generating 37.5 GWh of 89 electricity, that represents less than 1% of the total electricity produced per annum <sup>12</sup>. 90

91 The remaining biomass after AD, so-called digestate, can be further valorized as 92 organic fertilizer for crop cultivation, partly substituting mineral fertilizers <sup>13</sup>. In general, digestate is considered as an upgraded organic fertilizer since it is rich in 93 nitrogen. When digestates are applied according to best practice guidelines, that have 94 95 been recently researched and developed (such as better management and storage 96 conditions, i.e. storage facilities that are covered and/or have a high depth to surface area ratio) <sup>14</sup> they can be considered as an environmentally benign material <sup>15</sup>. Types of 97 digestate that are considered acceptable for use by organic farmers and growers are 98 listed in the EU regulation for organic farming <sup>16</sup>. In addition, in several countries, 99 especially in the UK, independent quality assurance schemes have been developed in 100 101 order to provide confidence to the market and the society that digestates are safe, 102 consistent and appropriate for use <sup>17–21</sup>. According to those schemes and regulations, 103 permitted waste input materials include wastes from dairy industry, such as materials 104 unsuitable for consumption or processing (solid and liquid dairy products, milk, food 105 processing wastes, yoghurt and whey) and biological sludge from on-site effluent 106 treatment. Anaerobic digestion, and further composting of the digestate, are currently 107 considered the most important technologies for the transformation of waste biomass 108 to biogas and nutrient recovery and account for up to 95% of biological treatment performed for organic waste <sup>22–24</sup>. 109

110 Uptodate, the majority of biogas plants are configured as single-stage installations. In this way, the microbial consortia that convert the biodegradable organic 111 matter to biogas are present within a single tank and operate under sub-optimal 112 conditions to achieve an overall balance between the sub-processes, i.e. hydrolysis, 113 114 acidogenesis, acetogenesis, methanogenesis. A variation of the traditional single-stage 115 configuration is the two-stage system in which two reactors are placed in series and 116 optimal trophic conditions are formulated for the distinct anaerobic microbial consortia. 117 Hydrolytic and acidogenic bacteria prevail in the first reactor, whereas methanogenic archaea dominate in the second one. Such configuration may a) produce hydrogen along 118 with volatile fatty acids in the acidogenic stage and increase the methane production in 119 the second (methanogenic) stage <sup>25</sup>, and b) avoid the imbalance caused by increased 120

121 acid production by the faster-growing acidogenic bacteria and the slower organic acid 122 consumption by the more sensitive methanogens, maintaining thus more favorable conditions for the different microbial groups <sup>26</sup>, among other advantages. Such a two-123 stage configuration may lead to increased energy production <sup>25</sup> due to the production 124 125 of hydrogen and methane blend and reduced key exhaust emissions when burning the 126 blend in an internal combustion engine compared with burning of methane alone <sup>27</sup>. 127 Although the two-stage anaerobic digestion systems seem to outmatch the conventional single-stage AD systems in various points it is still unclear if they will lead 128 129 to real environmental benefits. One way to investigate this is via Life Cycle Assessment 130 (LCA).

131 Several studies have been conducted focusing on the energy balances and 132 emissions of anaerobic digestion of various feedstocks, most notably studies by Styles et al.<sup>28</sup>, Fusi et al. <sup>29</sup>, Lijó et al. <sup>30</sup>. However, relatively little environmental assessment 133 work has been carried out for two-stage biogas production processes. Patterson et al., 134 135 compared the environmental burdens of a single-stage biogas (methane) production system against a two-stage (hydrogen/methane) production system using two 136 feedstocks with different characteristics and classifications. The systems boundaries 137 included raw biogas upgrade and its utilization as a vehicle fuel. The study showed that 138 139 the two-stage process using both feedstocks leads to reduction of the fossil fuel (diesel) burdens compared to the single-stage treatment <sup>31</sup>. Isola et al. assessed the 140 environmental impacts of a portable two-stage AD system fed with a mixture of food 141 142 waste and cardboard. According to their results the biogas generation rates from the portable AD system were comparable to a conventional full-scale system, while the 143 144 biogas combustion impacts were more sustainable compared to those associated with 145 conventional fossil fuels <sup>32</sup>.

Under the framework of LIFE10 ENV/CY/000721 project (Acronym: DAIRIUS) a
methodology has been developed in lab and pilot (demonstration) scale, for the
integrated management of EoL-DPs in Cyprus. The methodology included the
collection and transportation of EoL-DPs in a centralized biogas plant where EoL-DPs
were co-treated with agro-industrial wastes (AgW). Valorization scenarios of those

151 residues, that were examined in the present study, regarded their anaerobic co-152 digestion using a two-stage process realized in Continuous Stirred Tank Reactors 153 (CSTR), where EoL-DPs were acidified in a CSTR reactor, prior to their mixing with AgW in a methanogenic CSTR. In addition, co-digestion of EoL-DPs with AgW, in a single-154 155 stage CSTR was also investigated. The two systems were comparatively tested for a period of 9 months under pilot-scale conditions <sup>25</sup> and the environmental performance 156 157 of the processes which was assessed using a gate-to-gate LCA methodology is presented in this work. 158

159

#### 160 **EXPERIMENTAL**

#### 161 Pilot plant configuration

162 The pilot-scale experimental setup consisted of two conventional CSTR reactors, constructed by stainless steel, with 0.09/0.2 m<sup>3</sup> (acidogenic-CSTR) and 1.8/2.0 m<sup>3</sup> 163 164 (methanogenic-CSTR), working and total volume respectively. Both reactors were 165 periodically agitated, with a time-scheduled ON/OFF mode. The pilot plant comprised also of two stainless steel stirred feeding tanks with 0.2 m<sup>3</sup> total volume, one for the 166 agro-industrial wastes (AgW) mixture and the other one for the EoL-DPs mixture. Both 167 the acidogenic and the methanogenic reactor were operated under controlled 168 169 mesophilic conditions ( $37 \pm 1$  °C). The system had been operating for a total period of 350 days in the premises of a full-scale biogas facility (1 MW<sub>el</sub>) co-digesting AgW in 170 171 Cyprus. The AgW feedstock used was the same for the full scale and the pilot plant 172 system. In the first operational phase, the system run in a two-stage mode, with the 173 acidogenic reactor fed exclusively with EoL-DPs. After acidification the acidified 174 mixture was mixed with agro-industrial wastes (AgW) and co-digested in the

175 methanogenic bioreactor. In the second operating phase, the system operated without 176 the acidogenic stage, in a single-stage mode. The mixture of raw EoL-DPs and AgW was directly fed and co-digested in the methanogenic bioreactor. Both systems were 177 178 operated at Hydraulic Retention Time (HRT) of 37 days with the EoL-DPs mixture 179 accounting for ~20% (w/w) of the total feeding stream. Further details on the systems specifications and their operating performance during co-digestion under the different 180 operating scenarios have been previously described and can be found in our recent 181 study <sup>25</sup>. 182

183

#### 184 LCA methodology

185 Life cycle assessment (LCA) is an internationally accepted methodology used to provide insight into the environmental consequences of a process <sup>33</sup>. Its aim is to holistically 186 evaluate the environmental consequences of a product system or activity, by 187 quantifying the energy and materials used, the wastes released to the environment, 188 189 and assessing the environmental impacts of those in terms of energy, materials and 190 wastes. The environmental analysis conducted in this work was carried out according to ISO 14040 guidelines and recommendations <sup>34</sup>. 191 192 This LCA study was focused on the evaluation of the two AD processes tested in 193 the LIFE+ DAIRIUS project, with a view to the optimum energy valorization of EoL-DPs. 194 In such a gate-to-gate LCA, the upstream and downstream processes were not taken 195 into consideration, whereas waste treatment and bioenergy production were the 196 fundamental parts in the assessment boundaries.

#### 198 Goal and scope

199 The goal of this assessment was to identify, analyze and compare the life cycle 200 environmental impacts from a full-scale anaerobic co-digestion plant (AD) fed with 201 AgW and EoL-DPs in a ratio of 80%-20% (w/w) operating in either a single- or two-202 stage mode. In the second case, the acidification of the EoL-DPs stream takes place in 203 an acidogenic reactor prior to its mixing with AgW and feeding to the methanogenic 204 reactor. The objective was to identify hotspots affecting the environmental load of a 205 biogas generation plant. The impacts caused by the two scenarios were analyzed, 206 including the ones avoided from the displacement of fossil fuels. Comparison of the 207 two processes was also performed, based on their environmental performance. By 208 determining the environmental load of biogas production from AD, it is possible to identify whether the processes have beneficial or detrimental effects on the 209 210 environment.

211 If not all of the Life Cycle Assessment (LCA) can be carried out on the full life 212 cycle (from cradle-to-grave), special attention should be given in the analysis of the 213 intermediate stages of a product's life (from cradle-to-gate or from gate-to-gate) <sup>35</sup>. 214 For this LCA study, the complete life cycle inventory of industrial scale biogas 215 production with EoL-DPs is unavailable at the early design stage, which makes the 216 partial LCA (from gate-to-gate and nearly gate-to-grave) appropriate and practical for 217 evaluating possible environmental impacts. In this gate-to-gate LCA, the upstream (i.e. 218 the stages of production, collection and transportation of AgW and EoL-DPs to the AD

219	plant) and downstream stages (final use of generated products, such as digestate) of
220	the process developed will not be considered unless otherwise mentioned. The
221	biomass processing and energy production was the fundamental parts in the
222	considered assessment boundaries.
223	
224	Key assumptions
225	The functional unit must represent the function (common reference unit) of the
226	options compared <sup>36</sup> . The main function compared in this study is the bioconversion of
227	waste matter (biomass) into biogas and liquid fertilizer using either a single- or two-
228	stage mode of operation in the anaerobic digestion plant. So, in our case, for all
229	processes and treatment scenarios assessed, 1 ton of raw biomass consisting of 80%
230	(w/w) AgW and 20% (w/w) EoL-DPs, was used as the functional unit. In all scenarios
231	studied in this LCA analysis, the system boundaries were drawn within the biogas plant
232	limits once raw AgW materials and EoL-DPs were delivered to the plant. The data
233	obtained by the pilot plant operation were of vital importance. Based on those data,
234	the realistic energy requirements of such a system and the physicochemical
235	characteristics of the outputs were determined.

The present assessment examined the use of generated biogas for electricity and thermal energy production. Electricity was considered to be directed to the grid and consumed at the vicinity of the plant (gate-to-grave approach) ignoring thus any losses in the electricity grid due to distribution, whereas thermal energy was only used to cover the plant's own needs. However, AgW production and transportation to the plant, supply of the feedstock to the plant, transportation of the EoL-DPs to the plant, 242 de-packaging and packages recycling, transportation and distribution of the digestate 243 were not included in this LCA, since the main target of this work was to compare the two waste treatment scenarios. Possible methane emissions from manure storage on 244 245 the total global warming potential (GWP) of the biogas system were not taken into 246 account due to the fact that feedstock was used directly for feeding in the system. It 247 was also considered that the time needed for the various AgW to be treated via 248 anaerobic digestion is negligible compared to the timescale of environmental impacts. 249 Although the processing of anaerobic effluent (digestate) via centrifugation and the 250 subsequent treatment of the recovered solid fraction of digestate via aerobic 251 composting were considered as part of the system processes, and thus within the 252 system boundaries, the packaging of the produced compost and its distribution to the 253 market or direct spreading as a fertilizer was kept out (gate-to-gate approach). 254 However, it was assumed that the liquid fraction generated from digestate processing, 255 was directly spread in the surrounding area of the biogas plant facility for cultivation 256 purposes, avoiding thus any transportation (gate-to-grave approach). Alternative 257 processing of the liquid digestate fraction, such as aerobic or membrane treatment, 258 was not considered due to the complexity that would have been added to the scenarios compared in this study. The comparison of such alternative practices could 259 260 be the goal of another LCA and thus is considered to exceed the scope of the present 261 study, which mainly deals with the environmental assessment of the AD configurations 262 tested for the exploitation of the EoL-DPs.

263

#### 264 System description

265 Once agro-industrial wastes (i.e. 49% pig manure (PM), 14% liquid cow manure (LCM), 266 9% cheese whey (CW), 5% poultry wastes (PW) and 4% slaughterhouse wastes (SHW)) 267 and EoL-DPs (consisting of 93% milk, 5% yogurt, 2% white cheese) were collected, they 268 were transported to the main plant. In the first scenario, the EoL-DPs were acidified, 269 while simultaneous biohydrogen production was taking place (in an acidogenic CSTR 270 reactor under mesophilic pH-controlled conditions at pH 5.7±0.1) and after mixing with 271 the AgW were fed into the methanogenic mesophilic digester. On the other hand, in 272 the second scenario, the EoL-DPs were mixed with AgW and fed directly into the main 273 mesophilic digester. Recovered biogas from the bioreactor(s), containing carbon 274 dioxide and methane (methanogenic reactor) and hydrogen (in the case of two-stage 275 configuration), was burnt in a Combined Heat and Power (CHP) generator for the 276 production of electrical and thermal energy. The operating hydraulic retention time (HRT), in both methanogenic reactors, was considered to be the same (37 days), 277 278 simulating the operating conditions of the full-scale plant. The system boundaries of 279 the two bioprocesses are illustrated in Fig. 1 and Fig. 2, for scenario 1 and 2 280 respectively.

The system boundaries for both processes in this gate-to-gate LCA were defined from the physical limits of a typical centralized biogas plant, starting from the raw materials processing inside the facilities of the biogas plant including the energy production, the aerobic composting of produced digestate as well as the direct spreading and use of liquid digestate to adjacent arable land as water for irrigation. Only the inputs (e.g. raw materials, energy) and outputs (e.g. emissions) associated with the processes within the boundary limits were included. The inputs used for the

288	LCI database were the raw materials and energy needs, whereas outputs were the
289	emissions to the biosphere resulting from each process. Upstream activities (e.g.
290	animal breeding in cow farms, milk processing, cheese making, etc), transport and
291	downstream activities (e.g. distribution of the electrical energy to the grid, compost
292	packaging and usage) were not included within the boundaries of this study.
293	
294	Inventory data sources

- 295 Inventory analysis aims to quantify the inputs and outputs within the system
- boundaries. The result of an inventory is a long list of material and energy requirements,



**Figure 1.** System boundaries for an anaerobic co-digestion plant utilizing AgW and acidified EoL-DPs as feedstocks for biogas production in a two-stage process. Agro-industrial wastes (AgW) include 301 Slaughterhouse Wastes (SHW), Liquid Cow Manure (LCM), Cheese Whey (CW), Poultry Wastes (PW) and

302 Pig Manure (PM). CHP: Combined Heat and Power.



Figure 2. System boundaries for an anaerobic co-digestion plant utilizing AgW and EoL-DPs as feedstocks for biogas production in a single-stage process. Agro-industrial wastes (AgW) include Slaughterhouse

- 307 Wastes (SHW), Liquid Cow Manure (LCM), Cheese Whey (CW), Poultry Wastes (PW) and Pig Manure
- 308 (PM). CHP: Combined Heat and Power.

products and co-products as well as waste and outputs into the air, soil and water. This
list is referred to as the mass and energy balance or the inventory table. To establish a
life cycle inventory (LCI), the first phase is to survey and collect the life cycle data related
to the product system, from inputs to outputs. Life-cycle data concerning gaseous
emissions from biogas burning were obtained from a library of SimaPro 8.0.2 referring
to a 100 kW<sub>el</sub> (kilowatt electrical power) CHP engine having an electrical efficiency of
38% and a thermal efficiency of 46%.

317 LCI data were calculated on the basis of the functional unit of 1 ton of raw material entering the plant, and the energy needs for its treatment. For all processes, 318 319 the calculation of the energy needs and electricity production was carried out with the 320 hypothesis that all processes are carried out in Cyprus. Cyprus does not currently have 321 any primary energy sources and thus generation of electricity by the Electricity 322 Authority of Cyprus (EAC) is based exclusively on imported fuels, mainly crude oil. 323 Electricity production takes place in three power stations with a total installed capacity 324 of 1478 MW, as presented in SM Table 1.

325 The inputs into the AD process were the electricity use, for transferring wastes 326 between tanks within the facilities of the biogas plant, and stirring of different tanks 327 (i.e. mixing tank, acidogenesis and methanogenesis reactors, buffering and storage tank). The thermal energy required for heating the anaerobic digester(s) at mesophilic 328 329 conditions (i.e. 37 °C) and also for the pretreatment of SHW (80 °C for 2 hours) was a 330 fraction of the thermal energy recovered by the CHP unit after the combustion of the 331 produced biogas. Thus, external use of heat energy was not considered in the LCA, 332 since it was produced and consumed within the boundaries of the system.

The energy yields of the scenarios investigated in this study were based on calculations performed using results obtained from the demonstration pilot plant, which was operated in the framework of LIFE+ DAIRIUS project in Cyprus (see SM Table 2).

The energy equivalents used for the determination of the energy yields of the systems after combustion in a typical CHP generator are given in SM Table 3.

The energy requirements of the equipment of the system assessed and their operational period, by using a reference unit of 1 ton of treated effluent it is presented in SM Table 4.

342

#### 343 Impact assessment

344 Life cycle impact assessment is the phase where the results of the inventory analysis are 345 interpreted in terms of the impacts they have on the environment. The impact 346 assessments of the processes developed during LIFE+ DAIRIUS project were based on the internationally accepted ReCipe v.1.03. ReCiPe comprises a broadest set of endpoint 347 348 impact categories, including several environmental issues, to assess environmental 349 impact. Moreover, the results were simulated using the three different perspectives, 350 namely individualist (I), hierarchist (H) and egalitarian (E). The latter was finally chosen to evaluate the results, since it takes into account the long term, precautionary 351 environmental impacts, which better serve the scope of this study and thus the 352 353 following impact categories were identified: Climate change, Human health, Ozone 354 depletion, Human toxicity, Photochemical oxidant formation, Particulate matter

formation, Ionizing radiation, Climate change Ecosystems, Terrestrial acidification, Freshwater eutrophication, Terrestrial ecotoxicity, Freshwater ecotoxicity, Marine ecotoxicity, Agricultural land occupation, Urban land occupation, Natural land transformation, Metal depletion, Fossil depletion. Weighting of the results are also included in the present study which has been expressed by using the Pt value. Pt is a dimensionless value and each unit is equal to one thousandth of the yearly environmental load of one average European inhabitant.

362

#### 363 **RESULTS**

Based on the goal of this study, the hotspots of EoL-DPs and AgW treatments proposed here were identified. Moreover, the overall environmental performance of each

treatment scenario per ton of raw organic mixture entering the system, was quantified

367 and presented per impact category.

368

#### 369 **Overview of the results for Scenario 1 (two-stage system)**

370 The operating scenario of the two-stage process included the acidification of the EoL-

371 DPs in a mesophilic CSTR followed by their co-digestion with the AgW mixture. Under

the frame of this operating strategy a 31.1% overall increase in the energy yield of the

- 373 system was evident (SM Table 2). The main difference in the two operating scenarios
- 374 was the addition of an acidification step, and thus the supplementation of the LCI with

the relative energy inputs and outputs. More information regarding the systems'
performance in terms of biofuels production can be found in our recent study <sup>25</sup>.

377 The LCIA results of the process for the co-treatment of EoL-DPs with the 378 aforementioned AgW mixture, expressed per ton of raw biomass entering the plant, are presented in Fig. 3 (for details see SM Table 5). As can be seen, the environmental 379 380 performance of this scenario is generally affected by the composting process, the 381 application of the liquid digested matter to the land as fertilizer and the biogas 382 production stage, as a result of the atmospheric emissions generated during the 383 combustion of biogas in the CHP engine. The pretreatment stage had negligible effect 384 on the environmental performance of the system. The main inputs of the LCIA were 385 the electricity consumption due to the equipment used, while the main outputs were 386 the emissions ( $CO_2$ ) generated by the CHP engine during the combustion of the biogas and the biogenic emissions from the metabolic activity of the microorganisms during 387 388 composting. The use of digested liquid (anaerobic effluent) as fertilizer in agricultural 389 soil in the surrounding area of the biogas unit (without taking into account the 390 transportation of this liquid fertilizer) has also been part of this inventory.

Normalization is an optional step in LCA that is used for better understanding the relative importance and magnitude of the impact category indicator results<sup>37</sup>. Results obtained upon normalization are shown in Fig. 4. The most significant impact categories are shown to be human toxicity and terrestrial ecotoxicology of the liquid digested stream after its application as organic fertilizer. The rest of the parameters had negligible effect on the environmental parameters assessed.

- Based on the pilot plant results, the effect of the additional acidification step
- 398 on the energy consumption of the unit was negligible. So, the environmental
- 399 performance of such a plant was not affected as a result of the energy requirements of
- 400 the equipment used by the acidification stage.



402 Figure 3. Characterization data for Scenario 1 (two-stage system operation)



404

405 Figure 4. Normalization results for Scenario 1 (two-stage system operation)

In Table 1 the weighting of the impacts of Scenario 1 is shown. A total impact of
52.44 Pt is presented, while the disposal of the liquid digested stream is responsible for
68.82 Pt. In that Table the merits on the environment from the renewable energy
produced and the positive effect on fossil depletion are evident.

411

#### 412 Overview of the results for Scenario 2 (one-stage system)

413 The LCIA results of the process for the co-treatment of EoL-DPs with the AgW mixture, 414 expressed per ton of raw biomass treated in the plant, are presented in Fig. 5 and SM 415 Table 6. The environmental performance of this scenario is affected by the composting process (which was also the case for Scenario 1), the application of the liquid digested 416 417 matter to the land as fertilizer and the biogas production stage as a result of the 418 atmospheric emissions generated during biogas combustion in the CHP engine. Once 419 again, the pretreatment stage had negligible effect on the environmental performance 420 of the system. A positive effect is shown because of the energy recovery, both as 421 electricity delivered to the grid and thermal energy for covering the needs of the plant. 422 The main inputs of the LCIA were the electricity consumption of the pilot plant equipment while the main outputs were the emissions (CO<sub>2</sub>) generated by the CHP 423 424 engine during the combustion of the biogas and the biogenic emissions from the 425 metabolic activity of the microorganisms during composting. The use of digested liquid 426 as fertilizer in agricultural soil, and specifically in the surrounding area of the biogas 427 plant (without taking into account the transportation of the liquid fertilizer to the 428 agricultural soil), was also part of this inventory.

**Table 1.** Weighting of the impacts for Scenario 1 (two-stage system operation).

Impact category	Unit	Total	Feedstock storage	Pre- treatment	Acidogenesis	Methanogenesis	Post- treatment	Composting	Spreading of liquid anaerobic effluent to land	Electricity, with biogas engine	Electricity, production mix CY/CY U
Human Health	Pt	43.0765	0.1542	0.0074	0.0830	0.1055	0.1542	0.5540	49.9295	1.6260	-9.3775
Ecosystems	Pt	15.4701	0.0944	0.0045	0.0508	0.0646	0.0944	1.1347	18.6385	1.2239	-5.7378
Resources	Pt	-6.1056	0.1155	0.0055	0.0622	0.0790	0.1155	0.1028	0.25158	0.3052	-7.0231
Total	Pt	52.4411	0.3641	0.0174	0.1960	0.2491	0.3641	1.7916	68.8196	3.1552	-22.1384



**Figure 5.** Characterization data for Scenario 2 (one-stage system operation)

Whilst the characterization data show the relative contribution during each stage of the LCA, the characterization step does not show the relative significance of the impacts. Thus, a normalization step was undertaken, the results of which are shown in Fig. 6. The most significant impact categories were shown to be the human toxicity and the terrestrial ecotoxicology of the liquid digested matter after its application as organic fertilizer. The rest of the parameters had negligible effect on the environmental parameters assessed.

Fig. 7 illustrates the environmental merits of the process generated by the installation of an acidogenic reactor for the pretreatment of the EoL-DPs based on the weighting results of the processes.

The weighting of the impacts for Scenario 2 is presented in SM Table 7 and a total impact of 57.13 Pt is illustrated. The disposal of the liquid digested matter is responsible for the 68.82 Pt. Moreover, as in the case of Scenario 1, the environmental advantages associated with the biogas produced and the positive impact on fossil depletion are also presented.

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#### 451 **DISCUSSION**

In the present study, an analysis was conducted to determine the environmental performance of two integrated waste management processes for the valorization of EoL-DPs for bioenergy production, developed under the framework of LIFE+ DAIRIUS project. The main objective was the identification of the environmental hotspots of each operating scenario of EoL-DPs treatment, in order to provide



459 Figure 6. Normalized data for Scenario 2 (one-stage system operation)



**Figure 7.** Graphical representation of the assessment and the environmental effect by

- the EoL-DPs acidification.

feedback and support the sustainable development of these processes, as well as
future ones, in full-scale. The proposed plant was examined as a gate-to-gate
assessment.

472 According to the results obtained from this gate-to-gate LCA study it was 473 evident that prior acidification of EoL-DPs, followed by co-digestion with AgW 474 (Scenario 1, two-stage system), showed a better environmental performance 475 compared to the results obtained upon direct co-digestion in a mesophilic digester, 476 having a total impact of 52.44 Pt (Table 1) against 57.13 Pt (SM Table 7) respectively. Biogas production, and therefore energy yield, was higher for Scenario 1, reaching up 477 478 to 22.88 m<sup>3</sup> CH<sub>4</sub>/ton of feed (229.25 kWh/ton of feed), compared to Scenario 2 where biogas production was 17.45 m<sup>3</sup> CH<sub>4</sub>/ton of feed (174.85 kWh/ton of feed). This is the 479 480 main reason why the environmental performance of Scenario 1 was better than the 481 one of Scenario 2.

482 Weighting of the impacts for each category assessed in this study, including human health, ecosystem and resources, showed that the additional acidogenesis 483 484 stage in Scenario 1 had a slim contribution on the total negative environmental impact, 485 accounting only for up to 0.26%. Categories with negative impacts on the environment 486 mainly result from the combustion process of the biogas in the CHP generator, which produces gaseous emissions, and the electrical energy demands for its operation. 487 488 Therefore, air emissions, energy and thermal inputs during processing are the key 489 contributors to the environmental impacts in this LCIA.

Our results are in agreement with other LCA studies reported in literature. For 490 491 example, in a study where the environmental impacts of milk production in a dairy 492 farm located in Northern Italy were assessed, three scenarios were compared 493 regarding manure management, including: a) its storage in an open tank and 494 subsequent use as fertilizer, b) its anaerobic digestion for biogas production and heat 495 generation through biogas combustion and c) a scenario similar to (b) but the digestate was stored in a gas-tight tank <sup>38</sup>. It was found that for scenario (a) the GHG emissions 496 were 1.21 kg CO<sub>2</sub> eq.kg<sup>-1</sup>, whereas for scenario (b) and (c) the GHG emissions were 497 reduced to 0.92 (-23.7%) and 0.77 (-36.5%) kg CO<sub>2</sub> eq.kg<sup>-1</sup> respectively. However, for 498 499 cases (b) and (c) environmental impacts such as acidification, particulate size matter 500 emissions and photochemical ozone formation potential increased due to emissions 501 generated form the CHP engine.

502 In general, liquid effluents are stored for prolonged periods in anaerobic 503 lagoons before the final land application. In our study, the liquid digestate was directly spread to land without extended storage in anaerobic lagoon avoiding thus any 504 505 negative environmental impacts due to such storage. However, the use of the liquid 506 effluent (digestate) for cultivation purposes greatly contributes to the negative impacts 507 of the plant operation. The environmental impact of the liquid effluent application to 508 land was found to be 68.82 Pt in both cases. In particular, it was found that it had a 509 very significant impact on human health and the ecosystem in both scenarios. In the present gate-to-gate LCA study it was found that the application of the liquid effluent 510 511 to land contributed for up to 91.81% and 92.98% of the total negative environmental 512 impact for Scenario 1 and 2 respectively. Nevertheless, the anaerobically digested

513 liquid effluent still contains increased amounts of organic compounds (mostly 514 recalcitrant ones) and nutrients which are essential for cultivation purposes and can therefore replace chemical fertilizers. However, in this study, the positive effects due 515 516 to replacement of chemical fertilizers were not examined in detail because of the type 517 of analysis carried out (gate-to-gate). In this sense, a higher environmental gain would 518 have been achieved in this study by considering further processing of the digestate 519 rather than directly spreading it to land. Several digestate treatment technologies that 520 are able to provide environmental gains may be applied to this end. A recent study has 521 examined in-detail digestate treatment by (a) drying and pelletizing, (b) composting, 522 (c) biological treatment combined with reverse osmosis and drying, (d) ammonia 523 stripping and drying, and compared the results obtained with the ones derived from the case of direct spreading of the digestate on land <sup>39</sup>. It was concluded that, 524 525 compared to spreading, all alternative scenarios were characterized by a significant 526 reduction in air emissions, namely ammonia. Moreover, it was observed that the increase in energy intensity associated with those conversion processes seems to be 527 528 marginal due to the environmental benefits derived from other environmental 529 dimensions. Another scenario that has been proposed in order to reduce the environmental impact of the spreading of the liquid effluent to land, is the growth of 530 algae, and therefore the production of lipid-rich biomass, since those effluents are rich 531 in nitrogen and phosphorus. A study performed by Coats and colleagues <sup>40</sup> has 532 533 demonstrated that a two-stage AD configuration coupled with algae production results 534 in reduced GHG emissions by 60% compared to a traditional anaerobic lagoon.

535 Biological treatment, including anaerobic digestion and composting, is one of 536 the most frequently used techniques for bio-waste management, currently. Anaerobic digestion is particularly suitable for wet bio-waste and is perceived as a process for 537 538 energy recovery, producing biogas for energy purposes. Biogas can significantly reduce 539 greenhouse gas emissions (GHG) when injected into the gas distribution grid. In 540 addition, the residue from the process, the digestate, can be composted and used for 541 similar purpose as compost, thus improving overall resource recovery from the waste. 542 In this study, the environmental performance of a two-stage (acidogenesis followed by 543 methanogenesis) compared to a single-stage anaerobic co-digestion process of EoL-544 DPs with AgW was assessed. Positive impacts were evident because of the 545 replacement of electrical energy in the grid and thermal requirements with electricity 546 and thermal energy produced *in situ* in the plant via biogas combustion. Based on the 547 LIFE+ DAIRIUS pilot plant results, the effect of the acidification stage on the energy 548 requirements of such a plant in this gate-to-gate system is negligible. However, the 549 overall energy efficiency, and as a result the environmental performance of the 550 system, is increased due to the increase of the biogas yield in the two-stage scenario. 551 Therefore, further verification of results is needed on the environmental performance 552 of such a system using inputs from a full-scale two-stage plant. The environmental 553 assessment of such a system should be extended to a Cradle-to-Grave analysis, as part 554 of future work.

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