

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

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, w/w, ratio) was evaluated in a one-stage mesophilic anaerobic digestion (AD) process. 26 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 **RESULTS:** Prior acidification of EoL-DPs showed a better environmental performance compared to the results obtained upon direct co-digestion in a mesophilic digester, 30 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 m³ CH₄/ton of feed (229.25 kWh/ton of feed), compared to 17.45 m³ CH₄/on of feed (174.85 33 34 kWh/ton of feed) for the case where no pretreatment was performed. 35 **CONCLUSIONS:** The acidification of EoL-DPs enhanced the environmental performance of the process by reducing its impact by 8.2% (in Pt equivalents). The energy 36 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 process is required by extending the system's boundaries towards a Cradle-to-Grave 39 40 approach.

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

INTRODUCTION

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Nowadays, general scientific consensus believes that global warming is caused by the emission of anthropogenic greenhouse gases (GHG), mainly derived from fossil fuel combustion ¹. As a result, the demand for renewable energy is rising because of the increasing social awareness of consequences related to non-renewable energy use, e.g. fossil fuel depletion, energy security, and climate change (CC). Renewable energy production in the European Union is targeted to reach 20% and 27% of the total energy production by 2020 and 2030 respectively ^{2,3}. This transition requires insight into environmental alternatives of producing renewable energy, including CC, fossil fuel depletion, and land use changes. Bioenergy is a renewable form of energy produced from biomass, including energy crops, wood, microbial biomass as well as wastes from household, agriculture, cattle, forestry and industrial activities ⁴. Currently, there is a growing interest on the use of biomass for energy purposes in order to satisfy energy requirements all over Europe 5. Since biomass accounts for 2/3 of the renewable energy produced in Europe, its valorization results in lower dependency on fossil fuels for many European countries, depending on biomass local resources, in order to meet the renewable energy directive objectives ^{6,3}.

Biomass can be converted by anaerobic digestion (AD) into biogas, composed 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^3 , as in natural gas equivalent, whereas the projection for 2020 is about 28 billion m^3 . Biogas obtained can be exploited in situ to produce electricity or heat or preferably a combination of both through cogeneration in a combined heat and power (CHP) unit.

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 ^{7,3}.

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

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

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

acid production by the faster-growing acidogenic bacteria and the slower organic acid consumption by the more sensitive methanogens, maintaining thus more favorable conditions for the different microbial groups ²⁶, among other advantages. Such a two-stage configuration may lead to increased energy production ²⁵ due to the production of hydrogen and methane blend and reduced key exhaust emissions when burning the blend in an internal combustion engine compared with burning of methane alone ²⁷. 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 to real environmental benefits. One way to investigate this is via Life Cycle Assessment (LCA).

Several studies have been conducted focusing on the energy balances and emissions of anaerobic digestion of various feedstocks, most notably studies by Styles et al. ²⁸, Fusi et al. ²⁹, Lijó et al. ³⁰. However, relatively little environmental assessment work has been carried out for two-stage biogas production processes. Patterson et al., compared the environmental burdens of a single-stage biogas (methane) production system against a two-stage (hydrogen/methane) production system using two feedstocks with different characteristics and classifications. The systems boundaries included raw biogas upgrade and its utilization as a vehicle fuel. The study showed that the two-stage process using both feedstocks leads to reduction of the fossil fuel (diesel) burdens compared to the single-stage treatment ³¹. Isola et al. assessed the environmental impacts of a portable two-stage AD system fed with a mixture of food 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 biogas combustion impacts were more sustainable compared to those associated with conventional fossil fuels ³².

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

residues, that were examined in the present study, regarded their anaerobic codigestion using a two-stage process realized in Continuous Stirred Tank Reactors (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-stage CSTR was also investigated. The two systems were comparatively tested for a period of 9 months under pilot-scale conditions ²⁵ and the environmental performance of the processes which was assessed using a gate-to-gate LCA methodology is presented in this work.

EXPERIMENTAL

Pilot plant configuration

The pilot-scale experimental setup consisted of two conventional CSTR reactors, constructed by stainless steel, with 0.09/0.2 m³ (acidogenic-CSTR) and 1.8/2.0 m³ (methanogenic-CSTR), working and total volume respectively. Both reactors were periodically agitated, with a time-scheduled ON/OFF mode. The pilot plant comprised also of two stainless steel stirred feeding tanks with 0.2 m³ total volume, one for the agro-industrial wastes (AgW) mixture and the other one for the EoL-DPs mixture. Both the acidogenic and the methanogenic reactor were operated under controlled 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 MWel) co-digesting AgW in Cyprus. The AgW feedstock used was the same for the full scale and the pilot plant system. In the first operational phase, the system run in a two-stage mode, with the acidogenic reactor fed exclusively with EoL-DPs. After acidification the acidified mixture was mixed with agro-industrial wastes (AgW) and co-digested in the

methanogenic bioreactor. In the second operating phase, the system operated without 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 operated at Hydraulic Retention Time (HRT) of 37 days with the EoL-DPs mixture 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 operating scenarios have been previously described and can be found in our recent study ²⁵.

LCA methodology

Life cycle assessment (LCA) is an internationally accepted methodology used to provide insight into the environmental consequences of a process ³³. Its aim is to holistically evaluate the environmental consequences of a product system or activity, by quantifying the energy and materials used, the wastes released to the environment, and assessing the environmental impacts of those in terms of energy, materials and wastes. The environmental analysis conducted in this work was carried out according to ISO 14040 guidelines and recommendations ³⁴.

This LCA study was focused on the evaluation of the two AD processes tested in the LIFE+ DAIRIUS project, with a view to the optimum energy valorization of EoL-DPs. In such a gate-to-gate LCA, the upstream and downstream processes were not taken into consideration, whereas waste treatment and bioenergy production were the fundamental parts in the assessment boundaries.

Goal and scope

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

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

plant) and downstream stages (final use of generated products, such as digestate) of the process developed will not be considered unless otherwise mentioned. The biomass processing and energy production was the fundamental parts in the considered assessment boundaries.

Key assumptions

The functional unit must represent the function (common reference unit) of the options compared ³⁶. The main function compared in this study is the bioconversion of waste matter (biomass) into biogas and liquid fertilizer using either a single- or two-stage mode of operation in the anaerobic digestion plant. So, in our case, for all processes and treatment scenarios assessed, 1 ton of raw biomass consisting of 80% (w/w) AgW and 20% (w/w) EoL-DPs, was used as the functional unit. In all scenarios studied in this LCA analysis, the system boundaries were drawn within the biogas plant limits once raw AgW materials and EoL-DPs were delivered to the plant. The data obtained by the pilot plant operation were of vital importance. Based on those data, the realistic energy requirements of such a system and the physicochemical 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,

de-packaging and packages recycling, transportation and distribution of the digestate 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 the total global warming potential (GWP) of the biogas system were not taken into account due to the fact that feedstock was used directly for feeding in the system. It was also considered that the time needed for the various AgW to be treated via anaerobic digestion is negligible compared to the timescale of environmental impacts. Although the processing of anaerobic effluent (digestate) via centrifugation and the subsequent treatment of the recovered solid fraction of digestate via aerobic composting were considered as part of the system processes, and thus within the system boundaries, the packaging of the produced compost and its distribution to the market or direct spreading as a fertilizer was kept out (gate-to-gate approach). However, it was assumed that the liquid fraction generated from digestate processing, was directly spread in the surrounding area of the biogas plant facility for cultivation purposes, avoiding thus any transportation (gate-to-grave approach). Alternative processing of the liquid digestate fraction, such as aerobic or membrane treatment, 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 be the goal of another LCA and thus is considered to exceed the scope of the present study, which mainly deals with the environmental assessment of the AD configurations tested for the exploitation of the EoL-DPs.

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System description

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

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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

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

Inventory data sources

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,

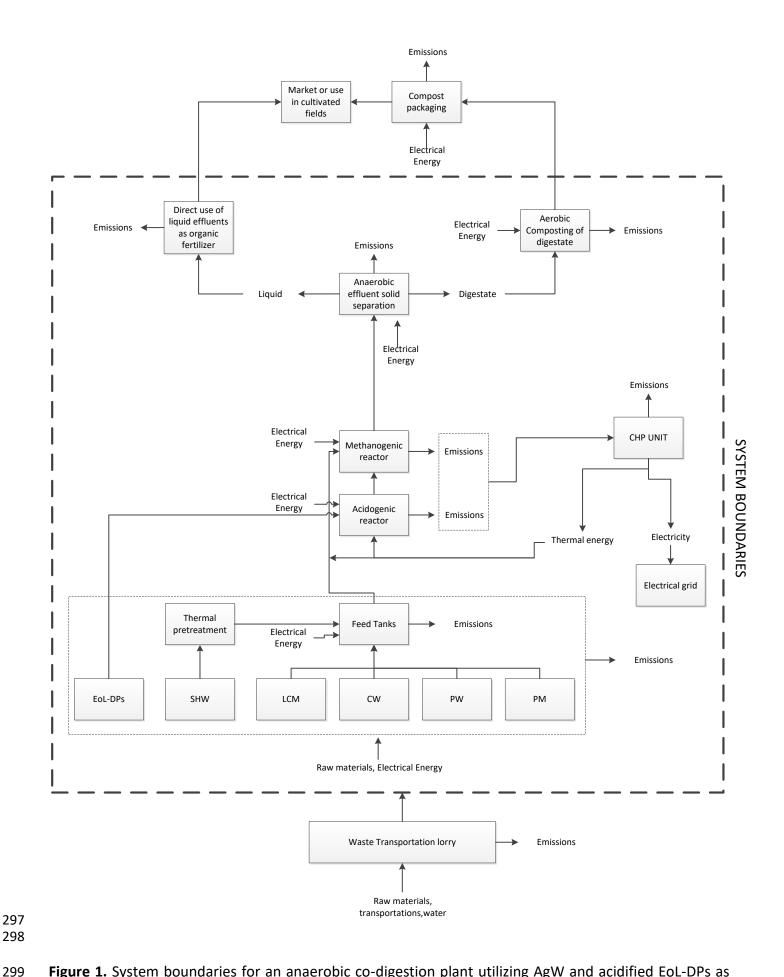


Figure 1. System boundaries for an anaerobic co-digestion plant utilizing AgW and acidified EoL-DPs as feedstocks for biogas production in a two-stage process. Agro-industrial wastes (AgW) include



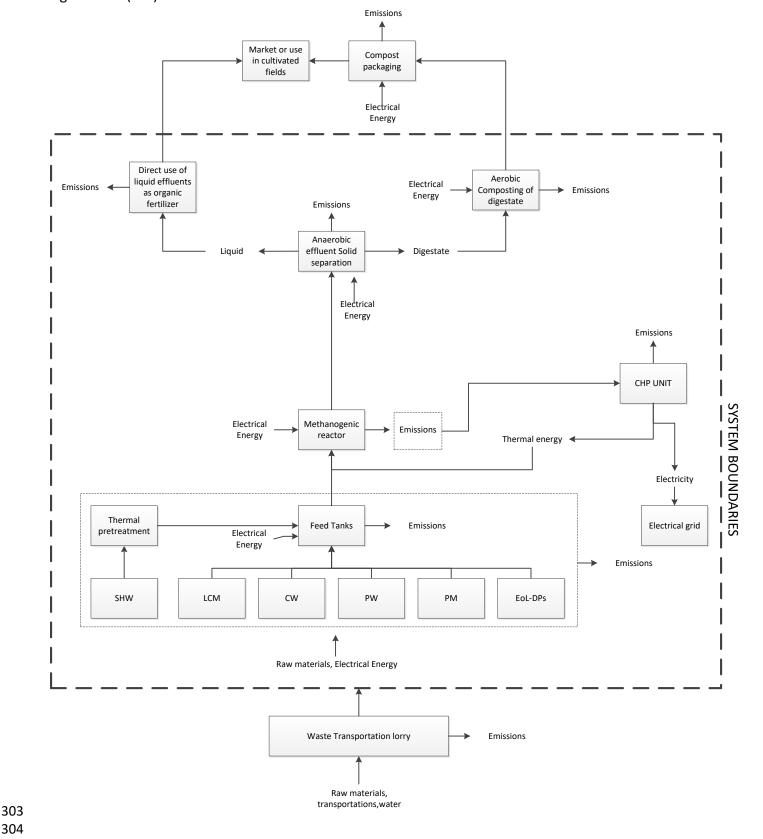


Figure 2. System boundaries for an anaerobic co-digestion plant utilizing AgW and EoL-DPs as feedstocks for biogas production in a single-stage process. Agro-industrial wastes (AgW) include Slaughterhouse

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

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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_{el} (kilowatt electrical power) CHP engine having an electrical efficiency of 38% and a thermal efficiency of 46%.

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, the calculation of the energy needs and electricity production was carried out with the hypothesis that all processes are carried out in Cyprus. Cyprus does not currently have any primary energy sources and thus generation of electricity by the Electricity Authority of Cyprus (EAC) is based exclusively on imported fuels, mainly crude oil. Electricity production takes place in three power stations with a total installed capacity of 1478 MW, as presented in SM Table 1.

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

Impact assessment

Life cycle impact assessment is the phase where the results of the inventory analysis are interpreted in terms of the impacts they have on the environment. The impact 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 impact categories, including several environmental issues, to assess environmental impact. Moreover, the results were simulated using the three different perspectives, 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 environmental impacts, which better serve the scope of this study and thus the following impact categories were identified: Climate change, Human health, Ozone 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.

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 and presented per impact category.

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

The operating scenario of the two-stage process included the acidification of the EoL-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 system was evident (SM Table 2). The main difference in the two operating scenarios 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 ²⁵.

The LCIA results of the process for the co-treatment of EoL-DPs with the 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 performance of this scenario is generally affected by the composting process, the application of the liquid digested matter to the land as fertilizer and the biogas production stage, as a result of the atmospheric emissions generated during the combustion of biogas in the CHP engine. The pretreatment stage had negligible effect on the environmental performance of the system. The main inputs of the LCIA were the electricity consumption due to the equipment used, while the main outputs were the emissions (CO₂) generated by the CHP engine during the combustion of the biogas and the biogenic emissions from the metabolic activity of the microorganisms during composting. The use of digested liquid (anaerobic effluent) as fertilizer in agricultural soil in the surrounding area of the biogas unit (without taking into account the 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 results³⁷.

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 397 on the energy consumption of the unit was negligible. So, the environmental performance of such a plant was not affected as a result of the energy requirements of 399 the equipment used by the acidification stage. 400

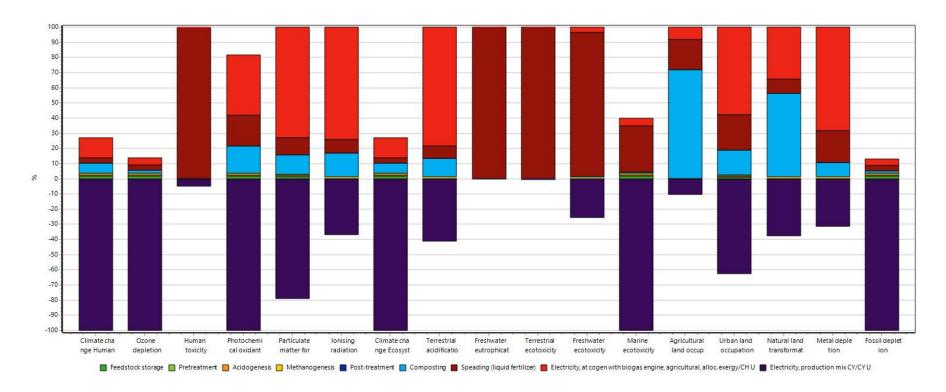


Figure 3. Characterization data for Scenario 1 (two-stage system operation)

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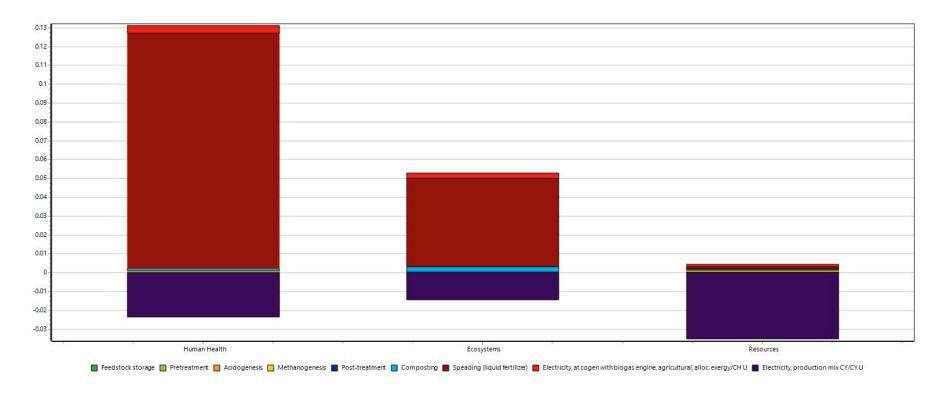


Figure 4. Normalization results for Scenario 1 (two-stage system operation)

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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.

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

The LCIA results of the process for the co-treatment of EoL-DPs with the AgW mixture, expressed per ton of raw biomass treated in the plant, are presented in Fig. 5 and SM 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 matter to the land as fertilizer and the biogas production stage as a result of the atmospheric emissions generated during biogas combustion in the CHP engine. Once again, the pretreatment stage had negligible effect on the environmental performance of the system. A positive effect is shown because of the energy recovery, both as electricity delivered to the grid and thermal energy for covering the needs of the plant. The main inputs of the LCIA were the electricity consumption of the pilot plant equipment while the main outputs were the emissions (CO₂) generated by the CHP engine during the combustion of the biogas and the biogenic emissions from the metabolic activity of the microorganisms during composting. The use of digested liquid as fertilizer in agricultural soil, and specifically in the surrounding area of the biogas plant (without taking into account the transportation of the liquid fertilizer to the 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

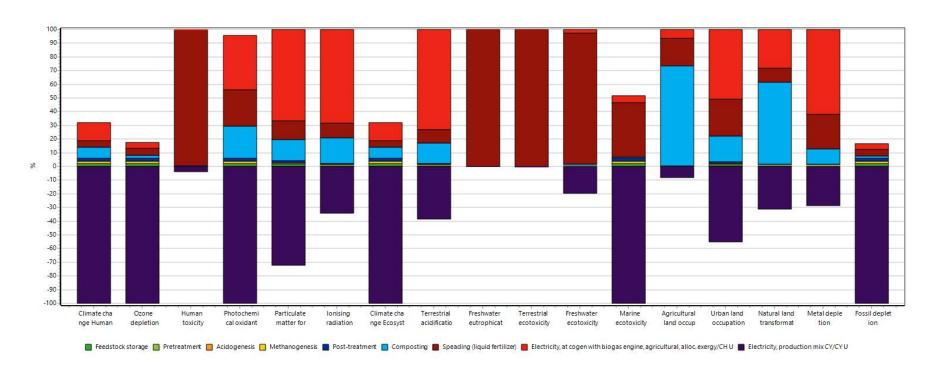


Figure 5. Characterization data for Scenario 2 (one-stage system operation)

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.

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

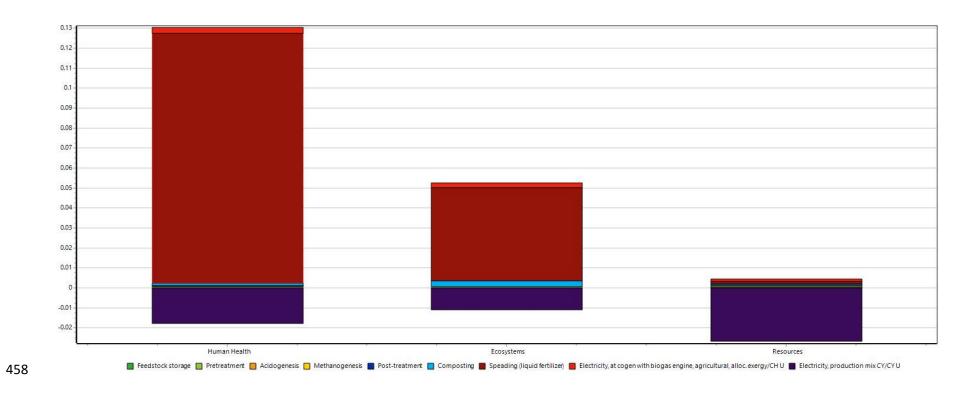


Figure 6. Normalized data for Scenario 2 (one-stage system operation)

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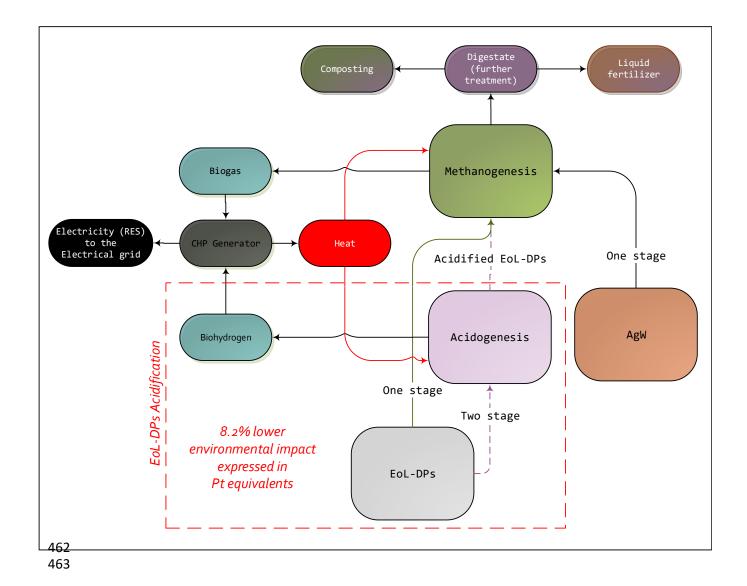


Figure 7. Graphical representation of the assessment and the environmental effect by the EoL-DPs acidification.

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.

According to the results obtained from this gate-to-gate LCA study it was evident that prior acidification of EoL-DPs, followed by co-digestion with AgW (Scenario 1, two-stage system), showed a better environmental performance compared to the results obtained upon direct co-digestion in a mesophilic digester, 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 to 22.88 m³ CH₄/ton of feed (229.25 kWh/ton of feed), compared to Scenario 2 where biogas production was 17.45 m³ CH₄/ton of feed (174.85 kWh/ton of feed). This is the main reason why the environmental performance of Scenario 1 was better than the one of Scenario 2.

Weighting of the impacts for each category assessed in this study, including human health, ecosystem and resources, showed that the additional acidogenesis stage in Scenario 1 had a slim contribution on the total negative environmental impact, accounting only for up to 0.26%. Categories with negative impacts on the environment 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.

Therefore, air emissions, energy and thermal inputs during processing are the key contributors to the environmental impacts in this LCIA.

Our results are in agreement with other LCA studies reported in literature. For example, in a study where the environmental impacts of milk production in a dairy farm located in Northern Italy were assessed, three scenarios were compared regarding manure management, including: a) its storage in an open tank and subsequent use as fertilizer, b) its anaerobic digestion for biogas production and heat generation through biogas combustion and c) a scenario similar to (b) but the digestate was stored in a gas-tight tank ³⁸. It was found that for scenario (a) the GHG emissions were 1.21 kg CO₂ eq.kg⁻¹, whereas for scenario (b) and (c) the GHG emissions were reduced to 0.92 (-23.7%) and 0.77 (-36.5%) kg CO₂ eq.kg⁻¹ respectively. However, for cases (b) and (c) environmental impacts such as acidification, particulate size matter emissions and photochemical ozone formation potential increased due to emissions generated form the CHP engine.

In general, liquid effluents are stored for prolonged periods in anaerobic 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 negative environmental impacts due to such storage. However, the use of the liquid effluent (digestate) for cultivation purposes greatly contributes to the negative impacts of the plant operation. The environmental impact of the liquid effluent application to land was found to be 68.82 Pt in both cases. In particular, it was found that it had a 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 to land contributed for up to 91.81% and 92.98% of the total negative environmental impact for Scenario 1 and 2 respectively. Nevertheless, the anaerobically digested

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

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

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REFERENCES

- González-García, S., García-Rey, D. & Hospido, A. Environmental life cycle
 assessment for rapeseed-derived biodiesel. *Int J Life Assess* 18, 61–76 (2013).
- 2. Office for the Official Publications of the European Communities. Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009.

 Official Journal of the European Union **52**, 16–62 (2009).
- 3. AEBIOM & European Biogas Association. A Biogas Road Map for Europe. 22
 (2009). Available at:
- http://www.aebiom.org/IMG/pdf/Brochure_BiogasRoadmap_WEB.pdf.
- Gerbens-Leenes, P. W., Hoekstra, A. Y. & van der Meer, T. The water footprint of
 energy from biomass: A quantitative assessment and consequences of an
 increasing share of bio-energy in energy supply. *Ecol. Econ.* 68, 1052–1060
 (2009).
- 5. Benoist, A., Dron, D. & Zoughaib, A. Origins of the debate on the life-cycle greenhouse gas emissions and energy consumption of first-generation biofuels a sensitivity analysis approach. *Biomass and Bioenergy* **40**, 133–142 (2012).
- 577 6. Summaries of EU legislation. Communication from the Commission to the

578 European Council and the European Parliament of 10 January 2007, 'An Energy 579 Policy for Europe'. COM2007, (2007). European Biogas Association. Green Gas Grids: Proposal for a European 580 7. 581 biomethane roadmap. (2013). Available at: http://european-biogas.eu/wpcontent/uploads/2014/02/GGG_European-Biomethane-Roadmap-final.pdf. 582 583 8. Cherubini, F. & Strømman, A. H. Life cycle assessment of bioenergy systems: 584 State of the art and future challenges. Bioresour. Technol. 102, 437–451 (2011). 9. 585 Fachagentur Nachwachsende Rohstoffe e.V. (FNR). Biogas – an introduction. 586 (2008). Available at: http://www.fabbiogas.eu/fileadmin/user_upload/Download/FNR_biogas_introd 587 uction.pdf. 588 589 10. Stambasky, J., Pfluger, S., Deremince, B. & Scheidl, S. Statistical Report of the European Biogas Association. (2016). Available at: http://european-590 591 biogas.eu/2015/12/16/biogasreport2015/. 592 Deremince, B. State of the Art and Future Prospects of Biogas and Biomethane 11. 593 in Europe. (2017). Available at: http://www.geotechnical.it/wp-594 content/uploads/2017/05/EBA-B.-DEREMINCE-CONVEGNO-BIOGAS-SOSTENIBILE.pdf. 595 596 Zachariadis, T. & Hadjikyriakou, C. State of the Art of Power Generation in 12. 597 Cyprus. in Social Costs and Benefits of Renewable Electricity Generation in 598 Cyprus 7–17 (SpringerBriefs in Energy, 2016). doi:10.1007/978-3-319-31535-5

- De Vries, J. W., Vinken, T. M. W. J., Hamelin, L. & De Boer, I. J. M. Comparing environmental consequences of anaerobic mono- and co-digestion of pig manure to produce bio-energy – A life cycle perspective. *Bioresour. Technol.*
- Whelan, M. J., Everitt, T. & Villa, R. A mass transfer model of ammonia
 volatilisation from anaerobic digestate. *Waste Manag.* 30, 1808–1812 (2010).
- Nicholson, F., Bhogal, A., Cardenas, L., Chadwick, D., Misselbrook, T., Rollett, A.,
 Taylor, M., Thorman, R., Williams, J. Nitrogen losses to the environment
 following food-based digestate and compost applications to agricultural land.
 Environ. Pollut. 228, 504–516 (2017).
- European Commission. Commission regulation (EC) No 889/2008. Official
 Journal of the European Union L 250, 1–84 (2008).
- Waste & Resources Action Programme (WRAP). Digestate and compost in
 agriculture. 1–32 (2016). Available at:
 http://www.wrap.org.uk/sites/files/wrap/Digestate compost good practice g
- uide_reference_version.pdf.
- Waste & Resources Action Programme (WRAP). Anaerobic digestate, Partial
 Financial Impact Assessment of the introduction of a Quality Protocol for the
 production and use of anaerobic degestate. WRAP Publications 44 (2009).

 Available at: http://www.organics-
- recycling.org.uk/uploads/category1060/Financial_impact_assessment_for_anae

- 620 robic_digestate.pdf.
- 621 19. Waste & Resources Action Programme (WRAP). Anaerobic digestate, End of
- waste criteria for the production and use of quality outputs from anaerobic
- digestion of source-segregated biodegradable waste. 1–27 (2014). Available at:
- https://assets.publishing.service.gov.uk/government/uploads/system/uploads/a
- ttachment_data/file/292473/426765_EA_QP_Anaerobic_Digestate_web.pdf.
- 626 20. Waste & Resources Action Programme (WRAP). BSI PAS 110 Specification for
- Digestate. (2012). Available at: http://www.wrap.org.uk/content/bsi-pas-110-
- 628 specification-digestate.
- 629 21. Waste & Resources Action Programme (WRAP). Using quality anaerobic
- digestate to benefit crops. 1–12 (2012). Available at:
- http://www.wrap.org.uk/sites/files/wrap/Using quality digestate to benefit
- 632 crops.pdf
- 633 22. European Commission. On the management of bio-waste in the European
- Union. Commission of the European Communities COM(2008), 18 (2008).
- 635 23. Jin, Y., Chen, T., Chen, X. & Yu, Z. Life-cycle assessment of energy consumption
- and environmental impact of an integrated food waste-based biogas plant. Appl.
- 637 Energy **151**, 227–236 (2015).
- 638 24. Oldfield, T. L., White, E. & Holden, N. M. An environmental analysis of options
- for utilising wasted food and food residue. *J. Environ. Manage.* **183**, 826–835
- 640 (2016).

- 641 25. Kopsahelis, A., Stavropoulos, K., Zafiri, C., Kornaros, M. Anaerobic co-digestion
- of End-of-Life dairy products with agroindustrial wastes in a mesophilic pilot-
- scale two-stage system: Assessment of system's performance. *Energy Convers.*
- 644 *Manag.* **165**, 851-860 (2018). doi:10.1016/j.enconman.2018.04.017
- 645 26. Ke, S., Shi, Z. & Fang, H. H. P. Applications of two-phase anaerobic degradation
- in industrial wastewater treatment. *International Journal of Environment and*
- 647 *Pollution* **23**, 65–80 (2005).
- Wang, J., Huang, Z., Fang, Y., Liu, B., Zeng, K., Miao, H., Jiang, D. Combustion
- behaviours of a direct injection engine operating on various fractions of natural
- 650 gas hydrogen blends. *International Journal of Hydrogen Energy* **32**, 3555 –
- 651 3564 (2007).
- 652 28. Styles, D., Mesa-Dominguez, E., Chadwick, D. Environmental balance of the UK
- biogas sector: An evaluation by consequential life cycle assessment. Science of
- 654 The Total Environment. **560-561**, 241-253 (2016).
- 655 *10.1016/j.scitotenv.2016.03.236*
- 656 29. Fusi, A., Bacenetti J., Fiala, M., Azapagic, A. Life Cycle Environmental Impacts of
- 657 Electricity from Biogas Produced by Anaerobic Digestion. Front. Bioeng.
- 658 Biotechnol. 4, 26 (2016). doi.org/10.3389/fbioe.2016.00026
- 659 30. Lijó, L., Lorenzo-Toja, Y., González-García, S., Bacenetti, J., Negri, M., Moreira, M.
- T. Eco-efficiency assessment of farm-scaled biogas plants, *Bioresour. Technol.*,
- **237**: 146-155 (2017). doi:10.1016/j.biortech.2017.01.055

- 662 31. Patterson, T., Esteves, S., Dinsdale, R., Guwy, A., Maddy, J. Life cycle assessment
- of biohydrogen and biomethane production and utilisation as a vehicle fuel.
- 664 Bioresour. Technol. **231**, 235-245 (2013).
- http://doi.org/10.1016/j.biortech.2012.12.109
- 666 32. Isola, C., Sieverding, H. L., Asato, C. M., Gonzalez-Estrella, J., Litzen, D., Gilcrease,
- P. C., Stone, J. J. Life cycle assessment of portable two-stage anaerobic digestion
- of mixedfood waste and cardboard. Resources, Conservation & Recycling 139,
- 669 114-121 (2018).
- 670 33. International Organisation for Standardisation (ISO). ISO-14040:2006.
- 671 Environmental Management Life Cycle Assessment Principles and
- Framework. (2006). Available at: https://www.iso.org/standard/37456.html.
- 673 34. Jiménez-González, C., Kim, S. & Overcash, M. R. Methodology for developing
- gate-to-gate Life cycle inventory information. *Int. J. Life Cycle Assess.* **5**, 153–159
- 675 (2000).
- 676 35. Kopsahelis, A., Kourmentza, C., Zafiri, C., Kornaros, M. Gate-to-gate life cycle
- assessment (LCA) of biosurfactants and bioplasticizers production via
- biotechnological exploitation of fats and waste oils. J. Chem. Technol.
- 679 Biotechnol. **93**, 2833-2841 (2018). doi:10.1002/jctb.5633
- 680 36. Leceta, I., Etxabide, A., Cabezudo, S., De La Caba, K. & Guerrero, P. Bio-based
- films prepared with by-products and wastes: Environmental assessment. J.
- 682 *Clean. Prod.* **64**, 218–227 (2014).

003	57.	neijungs, k., duniee, J., kleijii, k. & kovers, v. bias in normalization. Causes,
684		consequences, detection and remedies. Int. J. Life Cycle Assess. 12, 211-216
685		(2007). https://doi.org/10.1065/lca2006.07.260
686	38.	Battini, F., Agostini, A., Boulamanti, A. K., Giuntoli, J. & Amaducci, S. Mitigating
687		the environmental impacts of milk production via anaerobic digestion of
688		manure: Case study of a dairy farm in the Po Valley. Sci. Total Environ. 481,
689		196–208 (2014).
690	39.	Vázquez-Rowe, I., Golkowska, K., Lebuf, V., Vaneeckhautee, C., Michels, E.,
691		Meers, E., Benetto, E., Koster, D. Environmental assessment of digestate
692		treatment technologies using LCA methodology. 43 , 442–459 (2015).
693	40.	Coats, E. R. An integrated two-stage anaerobic digestion and biofuel production
694		process to reduce life cycle GHG. 459–473 (2013). doi:10.1002/bbb
695		
696		
695 696		