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1 **Environmental impacts of producing bioethanol and biobased lactic acid from**
2 **standalone and integrated biorefineries using a consequential and an attributional life**
3 **cycle assessment approach**

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10 **Abstract:**

11 This study evaluates the environmental impacts of biorefinery products using consequential (CLCA)
12 and attributional (ALCA) life cycle assessment (LCA) approaches. Within ALCA, economic allocation
13 method was used to distribute impacts among the main products and the coproducts, whereas within
14 the CLCA system expansion was adopted to avoid allocation. The study seeks to answer the questions
15 (i) what is the environmental impacts of process integration?, and (ii) do CLCA and ALCA lead to
16 different conclusions when applied to biorefinery?. Three biorefinery systems were evaluated and
17 compared: a standalone system producing bioethanol from winter wheat-straw (system A), a
18 standalone system producing biobased lactic acid from alfalfa (system B), and an integrated
19 biorefinery system (system C) combining the two standalone systems and producing both bioethanol
20 and lactic acid. The synergy of the integration was the exchange of useful energy necessary for
21 biomass processing in the two standalone systems. The systems were compared against a common
22 reference flow: “1 MJ_{EtOH} + 1 kg_{LA}”, which was set on the basis of products delivered by the system
23 C. Function of the reference flow was to provide service of both fuel (bioethanol) at 99.9%
24 concentration (wt. basis) and biochemical (biobased lactic acid) in food industries at 90% purity;
25 both products delivered at biorefinery gate. The environmental impacts of interest were global
26 warming potential (GWP₁₀₀), eutrophication potential (EP), non-renewable energy (NRE) use and
27 the agricultural land occupation (ALO). Regardless of the LCA approach adopted, system C
28 performed better in all impact categories than both standalone systems. The process wise
29 contribution to the obtained environmental impacts also showed similar impact pattern in both
30 approaches. The study also highlighted that the recirculation of intermediate materials, e.g. C₅ sugar
31 to boost bioethanol yield and that the use of residual streams in the energy conversion were beneficial
32 for optimizing the system performance.

33 **Keywords:** biobased products, attributional LCA, economic allocation, consequential LCA,
34 biorefinery, indirect land use change, system expansion

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1 **1. Introduction**

2 The increasing demand for biomass to biofuels has spurred the food vs fuels debates and has led to
3 investigate the impacts of devoting croplands for biofuels production (Lange, 2007; Marris, 2006).
4 Studies on 1st generation biofuel production (based on food crops) have stressed on their poor
5 environmental performance (Gressel, 2008; Mosier *et al.*, 2005; Sims *et al.*, 2010). Meanwhile, the
6 environmental life cycle impacts of the 2nd generation bioethanol production were also largely
7 determined by the types of biomasses and the system boundaries considered for the assessment (Luo
8 *et al.*, 2010). Example, switchgrass, sugarcane and sugar beet showed varied environmental
9 performance in the biofuel conversion pathway (Luo *et al.*, 2010; Muñoz *et al.*, 2013). Furthermore,
10 direct and indirect land use change (d/iLUC) impacts, as expected to be induced during the
11 production of biofuels and biobased products are also extensively debated (Khanna *et al.*, 2011;
12 Templer and van der Wielen, 2011). Moreover, biorefinery technologies are bringing new types of
13 biobased products (Cherubini, 2010) on a comparable functional basis to fossil based products
14 (Mickwitz *et al.*, 2011) and also aimed at addressing such environmental consequences by producing
15 both fuel and food/feed commodities. Maximizing the values of biomass feedstocks by utilizing most
16 of its components to produce both fuel and non-fuel products can be regarded as one of the
17 sustainable solutions to manage the available biomasses to meet the future multi-fold demand of
18 commodities (IEA, 2011; Parajuli *et al.*, 2015).

19 Among the different biorefinery concepts, the green biorefinery (GBR) technology is seen as an
20 alternative option for capitalizing the grassland biomass in Europe (Mandl, 2010; O’Keeffe *et al.*,
21 2011). The GBR, until now, primarily aimed at producing protein in order to reduce the import
22 dependency of livestock feed (e.g. soy cake and soy meal) and also producing high value chemicals
23 (e.g. lactic acid and lysine) (Kamm *et al.*, 2009). Green protein is important in the livestock sector,
24 whilst biobased lactic acid is important for the food, pharmaceuticals and chemical industries
25 (Ghaffar *et al.*, 2014; Kamm *et al.*, 2009; Kim and Moon, 2001; O’Keeffe *et al.*, 2011; Panesar *et al.*,
26 2007; Thomsen, 2004; Wee *et al.*, 2006). The global market production of biobased lactic acid in
27 2013 was 300-400 kilotons (ktons) (Harmsen *et al.*, 2014). The production is expected to reach 800
28 ktons in 2020 (Dammer *et al.*, 2013), driven by the demand of polylactic acid (Harmsen *et al.*, 2014).
29 In these contexts, biorefining of green biomasses is often seen as a sustainable path to deliver high
30 value biobased products and also achieving many societal goals (IEA, 2011). Despite its technical
31 viability are well described in many studies (Dale, 2003; Harmsen *et al.*, 2014; Kamm *et al.*, 2010;
32 Kamm *et al.*, 2009; O’Keeffe *et al.*, 2011), environmental impacts of its products’ value chains are
33 limitedly studied (Parajuli *et al.*, 2015).

34 Life cycle assessment (LCA) has been widely used as a tool for the assessment of environmental
35 performance of different products and services (European Commission, 2015a). According to ISO
36 (2006), the main phases of an LCA are (i) goal & scope definition: where the product or service to be
37 assessed is defined, a functional basis for comparison is chosen, (ii) inventory analysis: where the
38 details on the data used for the assessment are discussed, (iii) impact assessment: where the effects

1 of the resource use and the generated emissions are quantified into a limited number of impact
2 categories, and (iv) interpretation of the results: where results are reported in the most informative
3 way, along with the opportunities to reduce the impact of the product(s) or service(s). Furthermore,
4 whenever, a product system involves multiple products, choices on the approach to handle the co-
5 products are unavoidably connected (Thomassen *et al.*, 2008). With regard to the environmental
6 evaluations of different biobased products, it is thus relevant to develop and apply standardized LCA
7 methodologies that can cover the wide range of products delivered from a product system (European
8 Commission, 2015b; ISO, 2006). This is generally carried out by using either; sub-dividing the multi-
9 functional processes, system expansion and allocation (European Commission, 2010). In this
10 context, attributional (ALCA) and consequential (CLCA) approaches were aimed to resolve the
11 methodological debates over the allocation problems and also the choice of data (Thomassen *et al.*,
12 2008). Within ALCA approach, allocation can be avoided by using system expansion to handle the
13 co-products, but the co-product allocation is widely used (Thomassen *et al.*, 2008). In general, if
14 avoiding allocation is not possible, the ISO series (ISO, 2006) recommends using methods that
15 reflects the physical relationship, such as mass and energy content or using other relevant variables
16 to allocate, such as economic value of the products (Guinée *et al.*, 2004). In the current study,
17 economic allocation method was used, as is most frequently used (Crown and Carbon Trust, 2008).
18 Within CLCA approach, avoiding allocation by system expansion is the only acknowledged way to
19 deal with the co-products (Weidema, 2003). Moreover, it is also relevant to examine, whether the
20 choice of any of the methods would end-up with different conclusions on the environmental ranking
21 of any product system. Within such scope, comparative assessments using ALCA and CLCA approach
22 were also practiced in various studies, e.g. as reported in Thomassen *et al.* (2008) and Sanchez *et al.*
23 (2012).

24 This study aims at evaluating the environmental impacts of biorefinery products using a LCA
25 method. Evaluations were made for two standalone biorefinery plants, separately producing
26 bioethanol (system A) and biobased lactic acid (system B), and was compared with an integrated
27 system (system C) producing the both stated products. The integrated system was termed in
28 accordance to the definitions for “process integration” and “feedstock and product integration”
29 (Stuart and El-Halwagi, 2012). The integration aimed to assess possible synergies between two
30 different plants, so that they can be constructed at the same place to optimally utilize the resources
31 and minimize the related burdens of logistics. Evaluation was carried out by using both ALCA and
32 CLCA approach.

33 **2. Materials and methods**

34 **2.1 Goal and scope**

35 The goal of the current study is to evaluate and compare two standalone biorefinery systems with an
36 integrated biorefinery plant, which combine the two standalone systems on the basis of the possible
37 synergy between them. The study also examined whether CLCA and ALCA approach considered for
38 the environmental evaluation of biorefinery systems would arrive with same conclusions.

1 2.2 System boundaries, functional units and environmental impact categories

2 The evaluation covered the production and conversion of two different biomasses to produce two
3 biobased products in an integrated biorefinery system (system C). The assumed geographical
4 boundary was Denmark. A comparative assessment was made between system C and the two
5 standalone systems, and the evaluation is categorized into three scenarios. In the first scenario,
6 wheat straw is converted to bioethanol in a standalone plant (system A), while in the second scenario
7 alfalfa is converted to biobased lactic acid in another standalone biorefinery (system B). In the third
8 scenario, both bioethanol and biobased lactic acid are produced from wheat straw and alfalfa,
9 respectively, through the development of an integrated biorefinery plant (system C). In the
10 comparative assessment, such as the case discussed in this study, it is important to use the same
11 system boundaries and the same functional unit (FU) (EC, 2010). The system boundaries of the
12 evaluated systems are shown in Figs. 1-3. Combination of output products was one of the suggested
13 approaches to define FU in LCA studies related to biorefinery (Ahlgren *et al.*, 2015). Hence, to
14 compare system C with system A or system B, both products (bioethanol and biobased lactic acid)
15 produced from system C was considered as the main products. In this study, the FU was defined as
16 the production of a certain amount of bioethanol (@ 99.5% concentration) and biobased lactic acid
17 (with 90% purity) for use in transport and food industries, respectively. The reference flow was thus
18 a basket of (1 MJ_{EtoH} + 1 kg_{LA}), and the products delivered at the biorefinery gate. For a system that
19 generates only bioethanol or only biobased lactic acid, a 'conventional' standalone ethanol plant or
20 lactic acid plant is assumed to cover the ethanol or lactic acid deficit, respectively. In order words, if
21 the standalone systems are to be developed then the deficit products have to be supplied externally,
22 as shown in Figs 1-2. A similar approach was used to define FU, and was considered to compare
23 different agriculture systems (Marton *et al.*, 2016), bioenergy system (Djomo *et al.*, 2015) and for
24 evaluating biorefinery systems (Jungmeier *et al.*, 2013).

25 **Fig. 1.** Resource flow and system boundary of system A. Electricity produced represents net values
26 of the system (i.e., plant's own consumptions are subtracted). The dotted lines indicate the avoided
27 products considered in the CLCA approach.

28 **Fig. 2.** Resource flow and system boundary of system B. Electricity produced represents net values
29 of the system (i.e., plant's own consumptions are subtracted). The dotted lines indicate the avoided
30 products considered in the CLCA approach.

31 **Fig. 3.** Resource flow and system boundary of system C. Electricity produced represents net values
32 of the system (i.e., plants' own consumptions are subtracted). The dotted lines indicate the avoided
33 products considered in the CLCA approach.

34 The selected environmental impact categories were: Global Warming Potential (GWP₁₀₀),
35 Eutrophication Potential (EP), Non-Renewable Energy (NRE) use and Agricultural Land Occupation
36 (ALO). The first three impact categories were assessed using the "EPD" method (Environdec, 2013),
37 while the ALO was evaluated using the ReCiPe method (Goedkoop *et al.*, 2009). The selected

1 environmental impact categories were among the ISO preliminary list (ISO, 2006) and are relevant
2 whenever a production system and process are to be evaluated for minimizing agroecological
3 problems, such as induced due to GHG emissions, nutrient enrichments and resource use (fossil fuel
4 and land use). The selected environmental impact categories has also intended to consider both local
5 and global effects, in order to avoid the situation “the unintended increase of global impact is avoided
6 while trying to reduce local impact, or vice-versa” (van der Werf and Petit, 2002). Mitigation
7 opportunities to reduce GHG emissions implemented in one geographical setting can be regarded
8 useful to address the global climate change issues. Likewise, assessing eutrophication potential, a
9 regional or local effect (Smith *et al.*, 1999) helps to examine means of reducing nutrients losses and
10 emissions by management of resources, e.g. utilizing residual waste of biorefineries as fertilizer.
11 Likewise, impacts of fossil fuel depletion and occupation of agricultural land are relevant in the
12 current issues of resource use (Parajuli, 2016; Parajuli et al., 2015). The modelling for impact
13 assessment was facilitated by the use of the LCA software “SimaPRO 8.0.4” (PRé Consultants, 2015),
14 which incorporates the stated assessment methods.

15 2.3 Assessment approach

16 In the current study, after the basket of main products was defined for FU, other co-products were
17 handled by using both system expansion and allocation methods. Within system expansion, an
18 approach of CLCA was used to substitute the remaining co-products, as they have market values
19 (Table 2). Within ALCA, economic allocation method was used. Results obtained within CLCA and
20 ALCA approach for the identified biobased products were also compared under two criteria: first, to
21 determine if they consistently show similar impact pattern, e.g. on the environmental savings
22 compared to the conventional counterparts, differences on the net environmental impacts obtained
23 for the standalone systems and integrated system. The next criterion was the hotspot identification.

24 2.3.1 CLCA approach

25 In CLCA approach, whenever straw was involved as principal raw material to biorefinery, the
26 assessment included consequences of removing straw. Consequences were assessed relative to a
27 situation where straw is ploughed back to soil (Petersen and Knudsen, 2010). The consequences
28 were, in terms of: (i) emissions due to soil organic carbon (SOC) change (ii) compensation of
29 displaced nutrients by synthetic fertilizer and (iii) related N emissions avoided due to the straw
30 removal process. With regard to alfalfa, its environmental impacts were calculated based on the
31 input-output data, representative to the production from Danish arable land
32 (NaturErhvervstyrelsen, 2015; SEGES, 2010; SEGES, 2015). Data inputs for the biomasses
33 production are detailed in section 2.5.

34 Within CLCA approach, the identification of the main products (bioethanol and biobased lactic acid)
35 was based on the potential revenue generated from the products, calculated from the market price
36 and the produced mass of the selected biobased products (Figs. 1-3 and Table 2). The co-products

1 were assumed to partially substitute the corresponding marginal products (Figs.1-3). Marginal
2 products were assumed undergoing substitutions by the corresponding biobased products (Table 1).

3 2.3.2 ALCA approach

4 Within ALCA approach, the total environmental impacts of each biorefinery system were partially
5 allocated between its products and co-products using the method of economic allocation. Allocation
6 factors were calculated considering the market price of each product (Table 2) and the quantity of
7 products generated by each biorefinery systems (Figs. 1-3). A basket of two products (bioethanol and
8 biobased lactic acids) was again considered as the main products, and thus allocation factors were
9 estimated, which were based on the prices of each product. The calculated factors were: biobased
10 lactic acid plus bioethanol (70%) in system C; system A had 83% and system B (82%). Within ALCA
11 approach, the environmental impacts of the straw production was based on the study reported by
12 Parajuli *et al.* (2016).

13 **Table 1.** Biobased products and assumed substitutable products in the conventional market.

14 2.4 Basic assumptions and data sources

15 The basic assumptions related to this study are summarized in Table 2, unless otherwise are stated
16 in the text below. Life Cycle Inventory (LCI) of the product systems covered both; the background
17 and foreground processes (Figs. 1-3). The background process covered the environmental impacts of
18 producing materials and energy entering to the foreground level. Emission factors for the production
19 of assumed material inputs were based on Ecoinvent v3 (Weidema *et al.*, 2013). The foreground
20 process is the biorefinery system, covering the production and supply of the selected biomasses and
21 their conversion into the selected biobased products.

22 The materials and energy input entering to system A were: straw ((1 t, 85% dry matter (DM)),
23 enzyme, chemicals, water, heat and energy. The carbohydrate content of straw (system A) was
24 assumed as 76% (Møller *et al.*, 2005). Materials entering to biorefinery were calculated from the
25 studies reported by Bentsen *et al.* (2006), Kaparaju *et al.* (2009) and Wang *et al.* (2013). The mass
26 flow during the straw conversion to bioethanol is shown in the Supporting information/data (SI)-5
27 (Fig. S-5.1). Data on the potential environmental impacts (particularly, GHG emissions, EP and NRE
28 use) related to the production of enzyme (Cellic CTec3) was obtained from Novozymes (Kløverpris,
29 J.H, 2016, pers. comm.).

30 With regard to system B, the carbohydrate content of alfalfa was set to 56% (Møller *et al.*, 2005).
31 Crude protein (CP) and lactic acid content in the ensiled alfalfa were assumed as 15% and 6% of the
32 total DM biomass respectively (Møller *et al.*, 2005; O’Keeffe *et al.*, 2011) (Table 2). The dry matter
33 (DM) content of the harvested alfalfa was set to 35% (Møller *et al.*, 2005). It was assumed that alfalfa
34 is ensiled prior to its conversion to lactic acid, as ensiling is preferable for lactic acid production.
35 Ensiling also favours biomass storage without burden of drying the fresh green biomasses. It is also
36 useful to ensure a year-round supply of biomass to a biorefinery plant (Ambye-Jensen *et al.*, 2013).

1 The mass transformation of carbohydrate and CP contained in the fresh biomass to lactic acid and
2 feed protein respectively (Fig. 2) were partly based on O’Keeffe et al. (2011) (Table 2), and are
3 detailed in SI-6 (Fig. S-6.1). Unlike to O’Keeffe et al. (2011), the current study assumed enzymatic
4 hydrolysis to hydrolyse the press cake. Hydrolysis facilitates for an enhanced availability of glucose
5 from the carbohydrate contained in the pretreated biomass (Alvira *et al.*, 2010). This is useful to
6 increase the yield of biobased lactic acid than only depending on the juice fractions (see section
7 2.6.1).

8 **Table 2.** Basic assumptions on the parameters considered in the inventory analysis.

9 2.5 Life cycle inventory for the biomass production

10 Within CLCA approach, consequences of the straw removal process (see section 2.3.1) were
11 calculated for 1 ton (t) straw (85% DM). The consequences, in terms of emission due to SOC change
12 amounted to 143 kg CO₂ eq per t straw removed (Parajuli *et al.*, 2014). Additional processes, such as
13 baling, loading and the transport of straw to biorefinery plant were also considered to calculate the
14 total impact of utilizing the removed straw. Diesel consumed during baling, loading and transporting
15 the biomass (Table 3) was based on the study, reported by Dalgaard *et al.* (2001). Likewise, necessary
16 data for the alfalfa production were derived from Parajuli *et al.* (2017), and is reported in Table S-1.1
17 in SI-1.

18 Within ALCA approach, environmental impacts of producing straw from winter wheat was obtained
19 from Parajuli et al. (2016); and for alfalfa it was adapted from Parajuli et al. (2017).

20 A distance of 200 km (Bentsen *et al.*, 2009) single-trip was assumed for the transportation of both
21 biomasses to the biorefinery plant.

22 2.6 Life cycle inventory for biomass conversion

23 2.6.1 Standalone systems

24 *System A:* The basic data related to biomass production, particularly emissions are shown in Table
25 3. Likewise, energy and material inputs assumed for the biomass processing are also given in Table
26 3. Key stages for the straw conversion to bioethanol included: (i) pretreatment of the straw, (ii)
27 hydrolysis, (iii) fermentation, and (iv) recovery of the products. Hydrothermal pretreatment was
28 assumed, as it is suitable to breakdown lignocellulosic structures of straw into reactive cellulosic
29 intermediates (Galbe *et al.*, 2007). In the current study, after the hydrolysis process, the mass
30 transformation was assumed to follow a simultaneous saccharification and fermentation process and
31 then the distillation process (Galbe et al., 2007) . The total energy input, excluding the use of co-
32 produced energy was calculated as 0.98 MJ per MJ_{EtOH} (Bentsen et al., 2009), which was close to
33 the values reported in the studies, e.g. Pimentel and Patzek (2005) and Wang (2001). Likewise, the
34 direct primary energy input to the biorefinery plant (excluding the energy input to the biogas and
35 lignin fired combined heat and power (CHP) plants was calculated as 26 MJ per kg ethanol
36 production, which was also comparable with the range reported for cellulosic ethanol plant

1 (approximately 5-25 MJ per kg), as reported in the various studies (Kim and Dale, 2005; Luo *et al.*,
2 2009b; Pimentel and Patzek, 2005; Sheehan *et al.*, 2003). The process-wise contribution to the total
3 energy input was assumed as: pretreatment and hydrolysis (70%), saccharification and fermentation
4 (3%), product-recovery (24%). Generally, by-product recovery (including pelletization of lignin)
5 contributes about 10% of the primary energy input (Luo *et al.*, 2009a).

6 Potassium chloride (KCl) produced during the pretreatment of straw was considered to be recovered
7 as fertilizer (Larsen *et al.*, 2012) (Table 3) (Fig. 1). The recovery rate of KCl was set to 90%, and
8 equivalent amount of potassium was estimated using the ratio of molar weight of K to KCl. The solid
9 particle (lignin) collected from the distillation column was assumed to be fired in a CHP plant
10 (Eriksson and Kjellström, 2010; Nunes *et al.*, 2014). The CHP plant was also assumed to be
11 developed within the same location where biorefinery is assumed to be operated, in order to
12 minimize the logistic emissions. The liquid particles from the stillage and hydrolysate were
13 considered as substrates to produce biogas (SI-2 and SI-5). The energy balance showed that energy
14 recovery potential both from the biogas and lignin-fuelled CHP plants was able to fulfil 45% and
15 181% of the thermal and electric energy demand of the bioethanol system; hence heat energy
16 required was on deficit (Table 5).

17 **Table 3.** Primary input and output of materials related to the conversion of 1 t straw (with 85% DM)
18 to bioethanol (System A), all data are per 1 t straw.

19 *System B:* Table 4 summarizes the input-output flow of the materials and energy considered in
20 System B. The mass and energy flows for system B were calculated partly following the studies
21 reported by O’Keeffe *et al.* (2011) and Kamm *et al.* (2009). In the current study, lactic acid was
22 assumed to be produced from the (i) juice fraction and (ii) glucose fractions produced from press
23 cake. The glucose fraction was estimated from the hydrolysed mass of the press cake (i.e. fiber
24 portion) (detailed in SI-6). Sugar fractions in the press cake was assumed to be 33% (Cybulska *et al.*,
25 2010) of the total fiber fractions (on DM basis) (Fig S-6.1). The rest of the fiber particle contained in
26 the press cake was considered to be suitable as livestock feed (Kamm *et al.*, 2009), hereafter referred
27 as “fodder silage” (Table 4). Glucose produced after the enzymatic hydrolysis was thus considered
28 for the fermentation process. Extraction efficiency of crude lactic acid contained in the press juice
29 (i.e. 26 kg lactic acid per t DM juice) to produce the final product was set to 70% (O’Keeffe *et al.*,
30 2011), yielding 18 kg DM of biobased lactic acid. Likewise, the glucose to lactic acid conversion factor
31 was set to 79% (Doran-Peterson *et al.*, 2008), yielding 72 kg_{LA} (DM) from the estimated glucose to
32 be contained in the solid fractions of hydrolysed particles (see Fig S-6.1). The total production of
33 biobased lactic acid was thus estimated to be 89 kg DM (Table 4, SI-6). The calculated yield of
34 biobased lactic acid was close to the estimates reported in Kamm *et al.* (2009) (approx. 83 kg per t
35 DM of the grass silage under similar DM content of the raw biomass). Detailed process, including
36 the recovery of biobased lactic acid is elaborated in SI-6. Furthermore, liquid residue produced from
37 the processing of biomass was considered as substrate for biogas production, which was 6% of the

1 mass of decanted juice (Table 2), and was close to the amount reported in Kamm et al. (2009).
2 Energy consumption for these processes is summarized in Table S-3.1.

3 **Table 4.** Input-output of materials for the conversion of 1 t DM alfalfa to biobased lactic acid (system
4 B), all data are per t DM of alfalfa.

5 2.6.2 Integrated system

6 *System C:* Mass and energy flows during the combined production of both bioethanol and lactic acid
7 in system C are detailed in SI-4, Fig. S-4.1. System C was designed by assembling the two standalone
8 systems. During the integration, the energy, which was reported to be deficit in the standalone
9 systems, was partially covered by exchanging from one system to another (Table 5). The produced
10 biobased products were the same, as collectively described for system A and system B. The difference
11 in the systems was in the form of surplus electricity, and it was 1.23 GJ_e, estimated from the system
12 integration, but the system was still deficit in term of heat energy (Table 5).

13 2.7 Life cycle inventory for the secondary processing

14 2.7.1 Energy balancing for the biorefinery systems

15 The secondary processing of biomasses included handling of the residual products. For system A,
16 lignin was assumed as a biofuel for the combustion in a CHP plant producing heat and power.
17 Emissions from the combustion was assumed similar to Danish coal based CHP plants (Danish
18 Energy Agency, 2012). Likewise, C₅ molasses and the liquid residues were assumed to be collected
19 from system A and were considered for the conversion to biogas. The mass of the substrates for
20 biogas conversion in system A was calculated based on the studies reported by Bentsen et al. (2009)
21 and Kaparaju et al. (2009). In the case of system B, the handling of residual stream was the
22 conversion of decanted liquid residues to biogas production. The fermentable substrate for system B
23 was based on the proportion of volatile substance (VS) in the decanted press juice (O'Keeffe et al.,
24 2011) (Table 2) (see SI-6, Fig. S-6.1). For the both standalone systems, total methane yield was
25 calculated after Pugesgaard *et al.* (2013) (see detail in the SI-2). The energy balance of the entire
26 systems is shown in Table 5; whereas specific input to system A and system B is shown in Table 3
27 and Table 4.

28 **Table 5:** Energy balance calculated for the biorefinery plants. The balance accounted all useful
29 energy consumption within the biorefinery systems.

30

1

2 2.7.2 Nutrient recovery

3 Recoverable nutrients from the biogas digestate was calculated in the form of total N, P and K content
4 (Drosg *et al.*, 2015) (Table 2). The total mass of digestate was calculated after subtracting the losses.
5 Loss of total solids (TS) content was assumed as 50% of the available mass generated after the
6 anaerobic digestion compared to the initial pre-digester level (Drosg *et al.*, 2015; Lebuf *et al.*, 2013).
7 About 40% of the recoverable N and 100% each for P and K was assumed to substitute the equivalent
8 amount of synthetic fertilizer (Hansen *et al.*, 2006). Likewise, recovery of K from KCL was also
9 considered for System A (Table 3). The recovery of nutrients in system C was thus of the total
10 nutrients calculated for the two standalone systems. The recovered nutrients contained in the
11 digestate were assumed to be transported back to the farmers' field, considering similar distance of
12 transporting the biomass to the biorefinery plant.

13 2.8 Assessments on indirect land use change effects

14 iLUC was considered as the upstream consequences of occupying a productive land in Denmark
15 during the biomass production. Within CLCA, consequently induced GHG emissions was calculated
16 covering both, (i) upstream consequences of occupying a productive land in Denmark to produce
17 alfalfa and (ii) avoided iLUC assumed to be occurring due to the substitution of the alternative
18 agricultural products, assumed to be displaced by the biobased products. For the first element, iLUC
19 factor of 1.73 t CO₂eq ha⁻¹y⁻¹ was considered (Schmidt and Muños, 2014). For the second element,
20 the avoided iLUC was calculated based on the equivalent amount of feed products displaced due to
21 the production of feed protein and fodder silage from system B (Fig. 2) and system C (Fig. 3) (see
22 Table 2 for the composition of protein and energy feed). Feed protein produced from system B and
23 system C thus resulted to avoid 0.04 and 0.1 ha of land, which was assumed otherwise would be
24 occupied elsewhere for the production of corresponding marginal crops (i.e. spring barely and
25 soymeal respectively) to maintain the equivalent demand (Fig. 2 and Fig. 3) (Dalgaard *et al.*, 2007).
26 GHG emission avoided due to such avoidance of the land occupation was based on the carbon
27 footprint results of barley and soybean, as suggested in (Dalgaard *et al.*, 2007). In addition, while
28 calculating avoided iLUC, other chain effects in the form of so-called "soybean loop" (Dalgaard *et al.*,
29 2007) was also considered. The chain effect was assumed to initiate with an argument that due to
30 co-produced feed protein less soybean meal will be produced. Hence, a reduction in the production
31 of soybean and also soy oil will occur (Schmidt and Brandao, 2013), and such loss is compensated by
32 increasing the production of marginal oil, which may turned out to be palm oil, and so on. The
33 induced GHG emissions from the "soybean loop" was calculated based on the estimates reported in
34 Dalgaard *et al.* (2007), but was in an opposite analogy, as the current study deals for a reduction in
35 the demand of soymeal due to co-produced feed protein (Figs. 2-3). The study hence had following
36 key assumptions:

- 37 • 1 kg decrease of soybean meal would decrease the production of soybean to 1.005 kg.

1 • It resulted to compensate the demand of soy oil by palm oil, and the added burden would be in
2 the form of producing 0.86 kg of fresh fruit bunches and avoiding 0.012 kg of spring barley.
3 GHG emissions related to fresh fruit bunches and spring barely covered in the “soybean loop” were
4 based on the same study (Dalgaard et al., 2007), which were respectively, 177 and 671 kg CO₂ per t
5 product.

6 Considering the above effects, the net iLUC in terms of GHG emissions was calculated to be 1.6 t CO₂
7 ha⁻¹y⁻¹ during the production of 89 kg_{LA} (co-producing 34 kg of feed protein and 261 kg DM fibers)
8 and other non-agricultural products. The result was used only for system B and system C, as they
9 involved the production of alfalfa and had the co-products, which were assumed to be displacing the
10 marginal feed products (Table 1). It should be noted that within CLCA approach, straw was treated
11 with no iLUC effect (Schmidt and Brandao, 2013), provided that it depends on the assumed reference
12 situation (see section 5.3.2).

13 Within ALCA approach, the iLUC factor was assumed to be 2.7 t CO₂ ha⁻¹y⁻¹, which was amortized for
14 100 years after the value (270 t CO₂ eq ha⁻¹), as reported in Fritsche *et al.* (2010). A different iLUC
15 factor was considered within this approach than that of in CLCA approach, and was mainly to avoid
16 the methodological collision, as can be caused by summing average and marginal effects (Creutzig *et*
17 *al.*, 2012). In the current study, within ALCA approach, the iLUC factor considered was based on
18 Fritsche et al. (2010) and it was argued that growing bioenergy crops in 1 ha of land would displace
19 1 ha of the previous production. They estimated the change in the production based on the exported
20 products relevant for the bioenergy sector, e.g. soy and palm oil (Fritsche et al., 2010). Uncertainties,
21 with regard to the approach considered during the inclusion of iLUC in the assessment of net GHG
22 emissions are further discussed in section 5.3.2.

23 2.9 Sensitivity analysis

24 Sensitivity analysis is aimed to cover the potential uncertainties in the results due to different
25 assumptions and the process configurations, as discussed below.

26 2.9.1 Avoided products scenario

27 *Marginal electricity:* The sensitivity analysis included natural gas as a marginal fuel for electricity
28 generation (Mathiesen *et al.*, 2009).

29 *Marginal energy-feed:* Grass silage was assumed as marginal energy feed in the sensitivity analysis.
30 It was assumed that the fibers obtained from system B can meet the equivalent feed energy demand,
31 which is conventionally available from grasses to livestock. Furthermore, in the current study, since
32 alfalfa was considered as the principal raw material, the conventional way of directly utilizing the
33 forage, as a source of animal feed can be partially changed. The change was assumed to be covered
34 from the supply of fodder silage, co-produced from systems C and B (Figs.2-3; Table 1). In this
35 scenario, the supply of biomass was assumed to be from a Danish farm.

36 2.9.2 Variation on the yield of bioethanol and lactic acid

1 *Utilization of C₅ sugar to boost bioethanol yield:* In the basic scenario, only the fermentation of C₆
2 sugars was assumed to produce bioethanol, and the C₅ sugar contained in the molasses was used
3 during the biogas conversion. Alternatively, in the sensitivity analysis, a simultaneous conversion of
4 both C₆ and C₅ sugars was considered to boost the bioethanol yield. This was in accordance to the
5 claims that bioethanol yield can be increased if both C₆ and C₅ sugars are simultaneously fermented
6 with the use of an advanced yeast technology (Inbicon, 2013; Losordo *et al.*, 2016). For this
7 consideration hemicellulose (i.e. 0.057 t DM per t DM straw, see SI-5, Fig. S-5.1), which was
8 estimated to be in the hydrolysate, was considered as the intermediate raw material for the
9 fermentation process. The calculated increase in the bioethanol yield was 14% compared to the yield
10 reported in the basic scenario (Table 3). For the estimation, only 88% of the sugar contained in the
11 hydrolysate was assumed to be utilized, and thereby yielding 0.38 g bioethanol per gram sugar
12 converted (Biswas *et al.*, 2013). Hemicellulose, since was assumed to be used as intermediate raw
13 material, the consequences was in the form of a reduced mass of substrate available for biogas
14 conversion. Eventually, it resulted to a reduction in the net energy production. The calculated
15 reduction in the electricity production was 18% compared to the basic scenario. The calculated values
16 for the increase in the bioethanol yield and the reduction in the electricity production were close to
17 the range of 20% to 31% and from 12% to 37% respectively (Losordo *et al.*, 2016). During the
18 environmental evaluations, the added function to system B, as discussed in section 2.2 was also
19 adjusted accordingly along with the varied bioethanol yield.

20 *Varying the yield of biobased lactic acid production:* The yield of biobased lactic acid was varied
21 from -10% to +10% compared to the yield considered in the basic scenario (Table 4). The lower range
22 was set approximately to match with the yield reported in Kamm *et al.* (2009). During the evaluation,
23 such changes in the yield were also considered on the part of the added function assumed entering
24 to system A (section 2.1).

25 2.9.3 Calculation of energy and mass based allocation factors

26 In this case, mass and energy-based allocation method was considered for calculating the allocation
27 factors (parameters considered for the estimation is shown in Table 2).

28 3 Results

29 Table 6 shows the results for the selected environmental impact categories. Both net and gross
30 values of the impacts are presented. Net and gross values are with and without avoided impacts
31 respectively.

32 3.1 Overview of the standalone and integrated systems

33 The characterized results obtained within CLCA and ALCA approach for the selected biobased
34 products are shown in Table 6. The results revealed that compared to system A and system B, system
35 C performed better in most of the environmental impact categories. Within CLCA and ALCA
36 approach, net GWP₁₀₀ calculated per FU for system C were 58% and 20% lower than system A (Table

1 6). Within CLCA approach, when iLUC was considered net GWP₁₀₀ per FU for system B and system
2 C was increased by 39% and 84% compared to the results without iLUC (Table 6). Within ALCA
3 approach, net GWP₁₀₀ (with iLUC) was higher by 2%, 7% and 12%, respectively in system A, system
4 B and system C.

5 With regard to eutrophication potential, within CLCA approach the impact for system C was 86%
6 lower compared to system A, whilst, it was 6% higher within ALCA approach (Table 6). Likewise,
7 NRE use calculated for system C was 66% and 32% lower than system A, respectively within CLCA
8 and ALCA approach. On contrary to other impact categories, for both approaches, agricultural land
9 occupation for system C was higher than system A (Table 6).

10 With regard to the comparison between system C and system B, similar characteristics were found;
11 revealing better environmental performance for system C. Results showed that within CLCA and
12 ALCA approach, net GWP₁₀₀ was respectively, 86% and 31% lower in system C than system B,
13 followed by EP (101% and 63%), NRE use (97% and 52%) and ALO (94% and 38%) (Table 6).

14 **Table 6.** Environmental impacts of producing bioethanol and lactic acid from the standalone plants
15 and from system C (FU is “1 MJ_{EtOH} + 1 kg_{LA}”). Negative values indicate the environmental impact
16 abatement potentials.

17 3.2 Comparison of the systems based on CLCA and ALCA approach

18 Environmental impacts computed using CLCA and ALCA approach were compared for each
19 biorefinery system under following criteria.

20 3.2.1 Environmental differences

21 Within CLCA approach, net GWP₁₀₀ calculated for systems A, B and C were, respectively 97%, 91%
22 and 98% lower than that of the ALCA approach. In the similar order of biorefinery systems, EP was
23 respectively, 97%, 110% and 100% lower in CLCA compared to ALCA approach, NRE use (97%, 78%
24 and 99% respectively), ALO (99%, 77% and 98%) (Table 6). These revealed that regardless of the
25 approach used, the relative differences in the impact categories in each individual system
26 consistently showed the similar impact pattern.

27 3.2.2 Hotspots identification

28 *Contribution from the biomass production process:*

29 Fig 4 shows the contribution of different value chains of biorefinery in the standalone and integrated
30 systems. Within CLCA and ALCA approach, the related added function contributed 36% and 30%
31 respectively of the gross GHG emissions and it was 38% and 21% in system B (Fig 4 and Table 6).
32 Within CLCA and ALCA approach, the contribution from the biomass production was respectively,
33 16% and 17% of the gross GWP₁₀₀ obtained in system A. The same value chain contributed 57% and
34 55% in system B, and in system C it was 46% and 60%, respectively within CLCA and ALCA approach
35 (Fig. 4). Related emissions related to the biomass production, e.g. N₂O and emission due to SOC

1 change are shown in Tables 3-4. Within CLCA approach, emission due to SOC change contributed
2 14% of gross GWP₁₀₀ (system A). Likewise, SOC change related to the production of alfalfa mitigated
3 0.32% of the gross GWP₁₀₀ obtained for system B (i.e. -0.02 kg CO₂ eq per FU). In system C, GHG
4 emissions due to net SOC change (i.e. considering both the loss of SOC during the straw removal and
5 the sequestration during alfalfa production) contributed 11% of the gross GWP₁₀₀. Despite straw
6 removal resulted to a loss in SOC, consequently the avoided N₂O emissions mitigated 2% of the gross
7 GWP₁₀₀ (in system A). Likewise, N₂O emissions estimated for the alfalfa production, however, added
8 0.11% of the gross impact obtained for system B. Whilst, since net N₂O emissions was negative (i.e.
9 was avoided) in system C, it resulted to mitigate 0.3% of its gross impact. On contrary, within ALCA
10 approach, mitigation of GHG emissions due to SOC change was 6% of the gross GWP₁₀₀ for system
11 A. Likewise, it mitigated 0.3% and 10% of the gross GHG emissions, respectively obtained for system
12 B and system C. In the same manner, contribution to the gross GHG due to N₂O emissions was 7%
13 in system A, and was respectively, 0.1% and 8.3% in system B and system C.

14 With regard to EP, in system A the contribution to the gross impact was 7% and 25% computed
15 within CLCA and ALCA approach respectively. It was 77% and 43% of the obtained impact for system
16 B, and in system C (47% and 914%) (Fig. 4).

17 In the same manner, the same value chain covered 2% and 20% of the gross NRE use obtained within
18 CLCA and ALCA approach respectively. It should be noted that within CLCA approach, straw
19 accounted only energy used for baling and transporting the removed straw (Table 3), whilst within
20 ALCA approach, diesel consumed during winter wheat production was economically allocated to
21 straw. In system B, the biomass production process contributed 66% and 64% of the gross impact
22 calculated within CLCA and ALCA approach respectively; and for system C (62% and 97%) (Fig. 4).

23 With regard to the contribution to the gross ALO, the contribution in the same order of the approach
24 used, was 0.02% and 74% (system A); system B (103% and 79%) and system C (87% and 99%) (Fig.
25 4).

26 Major differences in the relative contributions with respect to the gross impact obtained for system
27 A and system C, as discussed above was mainly due to assumptions made on the process of
28 harnessing straw in the considered LCA approaches. The contribution pattern in both approach,
29 however, in vast majority of the cases were consistent and it also revealed that biomass production
30 was one of the principal contributors to most of the selected impact categories.

31

1 *Contribution due to material inputs and biorefining processes*

2 The contribution due to material inputs and the processing of biomass in system A were 48% and
3 53% of the gross GWP₁₀₀ obtained within CLCA and ALCA approach respectively, in system B it was
4 4% and 24%, and system C (54% and 40%) (Fig. 4). Likewise, the contribution to the gross EP from
5 the stated process was 72% and 58% (system A), respectively computed within CLCA and ALCA
6 approach. In system B, it was 2% and 28%, and in system C it was 53% and 6% of the respective gross
7 impacts (Fig 4). The contribution to EP in system C though was significantly different in relative
8 terms, but the absolute values were close within both approaches (e.g. 0.0002 and 0.0003 kg PO₄
9 per FU).

10 With regard to the contribution to the gross NRE use, it was 29% and 32% of the gross impact
11 obtained for system A, as obtained using CLCA and ALCA approach respectively (Fig. 4 and Table
12 6). For the same impact category, in system B, it was 0.8% and 20% of the gross impact obtained
13 within CLCA and ALCA approach respectively; and for system C (38% and 3%). Contribution to the
14 gross ALO, in the same order was 102% and 26% (system A); system B constituted with 0.3% and
15 6%, and in system C it was 13% and 1% (Fig. 4).

16 **Fig 4.** Contribution of processes involved in the entire biobased products chains.

17 3.3 Co-products handling and the environmental impacts

18 Within CLCA approach, the selected biobased products were credited substantially in terms of their
19 environmental footprints (Fig. 4). Likewise, within ALCA approach, burdens were significantly
20 shared by the co-products with respect to the environmental cost they owe during the operation of
21 biorefinery systems (Table 7). The results showed that within CLCA approach the co-produced
22 products, such as electricity, fodder silage (fibers) and feed protein contributed the most to the net
23 avoided impacts. Likewise, they also shared the most of the burdens within ALCA approach, after
24 the impact was attributed to the main products. Example, in system C, the results on GWP₁₀₀ showed
25 that electricity, fodder silage, recovered nutrients and feed protein contributed 55%, 28%, 12% and
26 2% of the net avoided impact (Table 7). Likewise, within ALCA approach, electricity and feed protein
27 had GWP₁₀₀ as 0.55 and 0.44 kg CO₂ eq per FU (Table 7).

28 **Table 7.** Contribution patterns of the co-products for avoiding and sharing the environmental
29 impacts within CLCA and ALCA approach. Units per FU in each system are: net GWP₁₀₀ = kg CO₂ eq,
30 EP = kg PO₄ eq, NRE use = MJ eq, and ALO = m².

31 **4 Results of the sensitivity analysis**

32 4.1 Natural gas as a fuel source for marginal electricity production

33 Assuming natural gas as the marginal fuel for electricity production, net GHG emissions calculated
34 for system A, system B, and system C was respectively, 10%, 71%, and 51% higher compared to the
35 basic scenario (Table 6). On contrary, net NRE use obtained for all the systems ranged 1-5% lower
36 than the basic scenario (Table 8). Higher net GWP₁₀₀ in the case of selecting natural gas as the

1 marginal fuel for electricity production was due to less environmental impact being displaced
2 compared to coal. In the same manner, compared to coal, fossil fuel intensity is lower in conversion
3 cycle of natural gas to energy.

4 4.2 Grass silage as an alternative source of animal feed

5 It resulted to increase both GWP₁₀₀ and NRE use for system B and system C significantly compared
6 to the basic scenario (Table 8). This was as a result of lower avoided impact compared to the basic
7 scenario (Tables 6-7).

8 4.3 Variations in the yields of biobased products

9 With the assumption on the recirculation of C₅ sugars, bioethanol yield was increased by 14%. On
10 such, GWP₁₀₀ obtained per FU for system A and system C was lower by 12%-15% compared to the
11 basic scenario (Table 6). Similarly, compared to the basic scenario, NRE use was reduced by 12-32%
12 in the designed biorefinery systems (Table 8).

13 Likewise, with a 10% increase or decrease in the biobased lactic acid, there were no significant
14 changes in the impact for system A, but the impact of systems B and C were affected (Table 8). This
15 revealed that the designed system was more sensitive to the bioethanol conversion processes and its
16 yield. Likewise, the cost of importing the conventional lactic acid would bear a higher impact, if
17 standalone systems are to be promoted instead of system C.

18 **Table 8.** Results obtained from the sensitivity analysis. Units per FU of each system are: net GWP₁₀₀
19 = kg CO₂ eq and NRE use = MJ eq.

20 4.4 Variations in the allocation factors

21 The allocation factor computed for bioethanol within system A and system C, as was considered in
22 the basic scenario was 46% and 39% respectively (section 2.3.2). Likewise, the allocation factor for
23 biobased lactic acid, as considered in the basic scenario was 37% and 32% in system B and system C
24 respectively. Uncertainties with the allocation factors computed using economic allocation method
25 may prevail due to surges in the future prices of biobased products. Furthermore, using the energetic
26 allocation method, the allocation factor obtained for bioethanol was 87% in system A and it was 68%
27 in system C, which was after covering the energy values of biofuel, electricity and biobased lactic acid
28 (Table 2). Likewise, using mass-based allocation method, allocation factor for bioethanol was 94%
29 and 31% respectively, in system A and system C. Within the same method, biobased lactic acid
30 resulted with allocation factor of 15% in system B and system C. It revealed that allocation factors
31 vary as per the types of products and the determining properties (e.g. market price, energy and other
32 utilities) of the products. The drawback of using mass and energy-based allocation method was
33 however related to their limitation for not covering products with different physical units and
34 functions. Example, energetic products like fuel, electricity are ignored when mass allocation is
35 considered; and likewise biobased lactic acid, feed protein and fodder silage are potentially ignored
36 if energetic-allocation is considered. Similar issues are also highlighted in other LCA studies, e.g. in

1 Singh *et al.* (2010), whenever a production system involve different products. This is however guided
2 by the scope and purposes of the assessment, e.g whether it aims for examining the internal
3 improvements in the manufacturing process, and/or, for the external communication to the market
4 (Svanes *et al.*, 2011).

5 **5 Discussions**

6 Discussions are made firstly by comparing the results calculated for the individual product with the
7 other related studies and also outlining the research perspectives. To calculate the environmental
8 impacts of the individual product, the contribution of the added functions (Fig. 4) was subtracted
9 from the net impacts (Table 6). This was assumed to be similar to the cases of assessing the impact
10 of separately producing bioethanol and biobased lactic acid, as from system A and system B.

11 5.1 Bioethanol production

12 On a product basis, the obtained results were found comparable with other studies. Example, if
13 contribution of the added function was neglected, net GHG emissions for producing bioethanol
14 would be 0.08 kg CO₂ per MJ_{EIOH} (or 2.3 kg CO₂ eq per kg_{EIOH}). The contribution due to added
15 function in this case was 36% to the gross GWP₁₀₀ (Fig. 4). The result obtained for bioethanol was
16 within the range, as reported for the lignocellulosic bioethanol (-0.007 to 3.9 kg CO₂ per kg_{EIOH})
17 (Borrion *et al.*, 2012; Degussa *et al.*, 2006; González-García *et al.*, 2012; Morales *et al.*, 2015; Muñoz
18 *et al.*, 2013; Wang *et al.*, 2013). The variations on the impacts in these studies were mainly due to:
19 different biomass feedstocks and methods considered in the evaluation. Following the similar
20 assumption made for neglecting the impact of the added function, within CLCA and ALCA approach,
21 the biomass production contributed 24-25% of the gross GWP₁₀₀, which was reported to be 30-60%
22 in Wang *et al.* (2013). The contribution from the enzyme production as obtained for system A was
23 29% of gross GWP₁₀₀ (i.e. 32% of net GWP₁₀₀), which was reported 40%-60% of the net impact in
24 Wang *et al.* (2013). Likewise, net NRE use obtained for bioethanol (system A) would was 0.49-0.69
25 MJ eq per MJ_{EIOH}, which was reported ranging from 0.1 to 0.8 MJ/MJ_{EIOH} (García *et al.*, 2011;
26 Morales *et al.*, 2015).

27 In the current study, the net savings, in terms of GHG emissions due to bioethanol production was
28 74% compared to petrol. GHG savings for an identical biorefinery plant was in the range of 44% to
29 89% (Cherubini and Ulgiati, 2010; Michael *et al.*, 2012; Wang *et al.*, 2013), and the results were
30 varying accordingly with different biomasses and assumptions made on the system boundaries.
31 Likewise, the net savings, in terms of NRE use due to bioethanol production was 91% compared to
32 petrol. For corn based bioethanol production, the savings on NRE use through the utilization of
33 bioethanol (E-85) was 95% and was 102% for E-100 (Sheehan *et al.*, 2003).

34 Under the most favourable conditions, enzyme dosage can be reduced by 30-50% through recovery
35 process (Ramos *et al.*, 1993), and without compromising the glucose yields (Weiss *et al.*, 2013). The
36 significance of such prospect can be seen clearly credited to bioethanol, as contribution of enzyme to
37 the environmental impacts (as discussed earlier) can be minimized. Likewise, the current

1 development in the enzymes production is also expected to lower the GHG profile by 50-70%
2 compared to Cellic CTec3 (Kløverpris, J.H. 2016.pers.comm.). This can also be regarded as another
3 potential opportunity to further lower the environmental footprints of lignocellulosic bioethanol.

4 5.2 Biobased lactic acid production

5 If the added function was neglected in system B, within CLCA and ALCA approach, the obtained
6 net GWP₁₀₀ for the production of 1 kg biobased lactic acid was 0.24 and 3.08 kg CO₂ eq respectively.
7 Within the approach of system expansion, results on GHG emissions were found ranging from -0.6
8 to 2.7 kg CO₂ eq per kg_{LA} (Degussa et al., 2006; European Commission, 2016). Likewise, within
9 economic allocation method, Daful *et al.* (2016) reported that GWP₁₀₀ for the biobased lactic acid
10 production was 4.34 kg CO₂ eq/kg_{LA}. With regard to NRE use, it ranged from 3.5 to 20 MJ/kg_{LA} (on
11 a cradle to cradle basis), and 32-43 MJ/kg_{LA} (on a cradle to factory gate basis) (Degussa et al., 2006).
12 In another study, European Commission (2016) reported that NRE use ranged from 9 to 37 MJ per
13 kg_{LA}. In the current study, without the added function to system B, net NRE use would be 10 and 50
14 MJ/kg_{LA}, within CLCA and ALCA approach respectively. The minor differences in the results with
15 the reported studies, e.g. European Commission (2016) were partly due to different: feedstocks
16 (corn, sugarcane and corn stover) and the assumptions made on the evaluation approaches (system
17 expansion and economic allocation, indicating the lower and the higher values respectively for the
18 reported impact). Furthermore, in the current study, alfalfa, since is nitrogen fixing plant,
19 application of synthetic N-fertilizer was not considered (SEGES, 2010). This also resulted to reduce
20 GHG emission, particularly during the fertilizer production and the application (Parajuli et al., 2017).
21 Upon the comparison with the conventional lactic acid, the net savings in terms of GHG emissions
22 due to the production of biobased lactic acid was 97% and 36%, respectively within CLCA and ALCA
23 approach. Savings in terms of NRE use was 88% and 30% compared to conventional lactic acid.
24 Hence, both approaches yielded with the same conclusion in terms of lower environmental footprints
25 compared to their conventional counterparts.

26 5.3 Methodological dilemma and perspectives

27 5.3.1 Consequences of straw removal

28 In the current study, the consequence of removing straw, in terms of emissions due to SOC change
29 was calculated as 143 kg CO₂ eq per t (85% DM). With regard to the removed mass of straw, it was
30 31% of the total straw yield (Parajuli et al., 2016). The impact of such can be neglected if the concept
31 of “sustainable rate of residues recovery” is considered. Sustainable recovery rate was suggested to
32 be within the range of 33% and 50%, and is also argued for a nominal effect on SOC change (Scarlat
33 et al., 2010; Spöttle et al., 2013). Scarlat *et al.* (2010), while suggesting 40% as the recovery rate; also
34 argued that it can be sustainably removed from the field once every 2.5 years on average. This may
35 stress to examine the consequences to an agro-ecosystem when large and commercial scale
36 biorefineries, primarily depending on straw are to be developed, keeping in mind that there are other
37 sectors demanding the same biomass (Gylling *et al.*, 2013). Likewise, maintaining SOC, even

1 removing straw was argued could be only possible if the grain yield exceeds certain level (Johnson *et al.*, 2006; Tarkalson *et al.*, 2009). Considering the wide range of the applications of straw in different
2 sector (Gylling *et al.*, 2013), it might be relevant to assess the consequences of using straw with
3 respect to a reference situation that it may affect.
4

5 5.3.2 *iLUC and the system boundaries*

6 With regard to the impact induced due to iLUC, there are some studies urging for more scientifically
7 robust and consistent way of assessing the impact, if it should be included in carbon footprint
8 assessments (Finkbeiner, 2013). Furthermore, Langeveld *et al.* (2014) suggested that the evaluation
9 should be enriched by incorporating more information on land use changes and examine influences
10 of local cropping patterns, as well as differences in current and potential productivities in different
11 agro-ecologies and farming systems. It was further supported by another critic, “given that there is
12 no accepted approach to estimate the global effects of biofuel policy on land-use change, it is critical
13 to assess the actual effects of policies through careful analysis and interpretation of empirical data”
14 (Kline *et al.*, 2011). Despite the stated critics, a general consensus, particularly on the prevalence of
15 effect was also found (Gawel and Ludwig, 2011; Kløverpris and Mueller, 2013). Moreover, one of the
16 major challenges and debated issue is on the methods to quantify the induced GHG emissions and
17 the uncertainties associated with it (Broch *et al.*, 2013; Di Lucia *et al.*, 2012; Warner *et al.*, 2014).
18 Several methods have quantified the impact of iLUC, e.g. in Audsley *et al.* (2009), Cederberg *et al.*
19 (2011) and Schmidt *et al.* (2015). A generic iLUC factor of 1.73 t CO₂ eq ha⁻¹y⁻¹ (Schmidt and Muñoz,
20 2014) was used in this study. Moreover, the iLUC factor suggested in other studies are: 1.43 t CO₂ eq
21 ha⁻¹y⁻¹ (Audsley *et al.*, 2009) and 1.9 t CO₂ eq ha⁻¹y⁻¹ was suggested for the world average arable land
22 (Schmidt and Muñoz, 2014). Likewise, iLUC factors were also reported varying with the assumptions
23 on the types of the occupied land, e.g. for soy meal production it ranged from 1.5 to 10 t CO₂ ha⁻¹y⁻¹.
24 The highest value was reported if the conversion takes place in forest land and the lowest was in
25 grassland (Leip *et al.*, 2010). These showed that the results on GHG emissions of the biobased
26 products, including iLUC then would have higher degree of variations based on the factors assumed
27 in the calculation.

28 Likewise, in the current study through “soybean loop”, we assumed that during the avoidance of
29 soymeal the effect would be on the production of soy oil, and on such marginal oil has to compensate
30 the demand. It was assumed that palm oil was the marginal oil, however it is difficult to be certain
31 which of the oil types (palm or rape seed) would be the marginal source (Dalgaard *et al.*, 2007).
32 Likewise, within CLCA approach, the conversion of straw to bioethanol was regarded without iLUC,
33 as there are some claims inferring that it may depend on the designed scenarios of utilizing the
34 residual resources (Schmidt and Brandao, 2013). For instance, if C₅ molasses produced in system A
35 and system C was to be regarded as protein source, and assumed to displace soymeal then there
36 could be a state where other value chains are affected (Bos *et al.*, 2016; Schmidt and Brandao, 2013).
37 This effect, however, can be ignored in the current study, as the consequences were accounted with
38 respect to different reference situation of straw utilization. Other alternative scenarios for assessing

1 the consequences of straw could be on their feed values (Tonini *et al.*, 2016). Regardless of any
2 scenarios, most importantly double counting on any cases should be avoided (Finkbeiner, 2013;
3 Pawelzik *et al.*, 2013).

4 Furthermore, the iLUC factor assumed for ALCA approach is also uncertain along with the changes
5 in the future yield of crops (Finkbeiner, 2013), as the factor was found calculated based on the “global
6 mix” for agricultural exports (Fritsche *et al.*, 2010; Fritsche *et al.*, 2010). It is therefore important to
7 reach to a standardized methodology that can be applied for assessing iLUC factors and used for
8 calculating the carbon footprints of biobased products.

9 **6 Conclusions**

10 The current study highlights that the benefits of the system integration for bioethanol and biobased
11 lactic acid productions were in terms of higher net savings of GHG emissions, NRE use and EP
12 compared to the standalone systems. However, the obtained ALO was higher in the integrated
13 system than the standalone system. Based on the comparison of the results obtained within CLCA
14 and ALCA approaches, it can be concluded that the recommendations for producing biobased
15 products from an integrated system would be the same, regardless of the approach used. The two
16 approaches had similar impact pattern for most of the impact categories, e.g. as was revealed from
17 the environmental differences obtained from the comparison among the biorefinery systems, and
18 from the hotspots identification. Both bioethanol and biobased lactic acid had net environmental
19 gains compared to petrol and conventional lactic acid respectively, regardless of the approach used.

20 GHG emissions in agriculture stage were determined by the emission of nitrous oxide and due to
21 SOC change, whereas in biorefinery processes it was determined by emissions related to: energy
22 input and from the enzyme production. SOC change was important to partly mitigate the GHG
23 emissions in an integrated system.

24 The results also showed that the net avoided impact would vary along with the different assumptions
25 on marginal products. For example, system B and system C were with lower avoided impacts when
26 locally produced grass-silage was assumed as marginal source of energy-feed instead of Ukrainian
27 barley, as discussed in the sensitivity analysis. The study also showed that the benefits of
28 recirculating intermediate raw materials were optimize the performance of biorefinery and to reduce
29 the environmental footprints of biobased products. Example, the recirculation of C₅ sugar resulted
30 to increase the yield of bioethanol by 14%, which eventually reduced the GHG emissions
31 approximately by 12-29% compared to the case of fermenting only the C₆ sugar, as discussed in the
32 sensitivity analysis for systems A and C.

33 Finally, for the optimizations of biorefinery systems following resource integrations can be further
34 beneficial: (i) utilization of intermediate raw materials for producing biochemicals, e.g. in system A,
35 even after the enzymatic hydrolysis, the glucose can be partitioned to produce both fermentable
36 products, bioethanol and biobased lactic acid, (ii) recirculation of enzyme. Likewise, environmental
37 sustainability assessments of the selected biobased products in a commercial scale is inevitably

1 relevant to attract the investors and for formulating conducive policies. Last but not least, economic
2 evaluation of producing the biobased products is also relevant for systemic sustainability evaluations
3 of biobased products.

4 **Acknowledgement**

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13 Soil Science and Plant Cultivation, Poland, for her constructive suggestions to this study, provided
14 at the time the current study was initially presented.

15 **Appendix A:** Supplementary data

16 **Appendix B:** Graphical abstract

17

1 **Figure. captions**

2 **Fig. 1.** Resource flow and system boundary of system A. Electricity produced represents net values
3 of the system (i.e., plant's own consumptions are subtracted). The dotted lines indicate the avoided
4 products considered in the CLCA approach.

5 **Fig. 2.** Resource flow and system boundary of system B. Electricity produced represents net values
6 of the system (i.e., plant's own consumptions are subtracted). The dotted lines indicate the avoided
7 products considered in the CLCA approach.

8 **Fig. 3.** Resource flow and system boundary of system C. Electricity produced represents net values
9 of the system (i.e., plants' own consumptions are subtracted). The dotted lines indicate the avoided
10 products considered in the CLCA approach.

11 **Fig 4.** Contribution of processes involved in the entire biobased products chains.

12

13

1 **List of Tables:**

2 **Table 1.** Biobased products and assumed substitutable products in the conventional market.

| Biobased products and unit (kg) | Substitutable products for co-products scenarios and data sources |
|---|--|
| Bioethanol | Petrol |
| Lactic acid (kg) | Conventional Lactic acid: (GLO) market ^a . |
| Feed protein | Soybean meal: (GLO) market ^{a,b} |
| Fodder silage (mainly fiber-residues) (kg) | Ukrainian barley (Ukraine), as energy feed ^c ((data as: Gross (GLO) barley grain to generic market ^a)) |
| Electricity (kWh) | Coal fired electricity production, DK ^{a, d} . |
| Digestate (kg) ^e | Recovered from the designed systems (Figs. 1-3) |
| Added functions | Conventional lactic acid (System A) ^f Bioethanol based on biomass (System B) ^g |

Assumptions:

^a Database for CLCA and ALCA approach adapted from Ecoinvent v3 (Weidema *et al.*, 2013).

^b Crude Protein (CP) for feed protein = 65% CP (O'Keeffe *et al.*, 2011). Soybean meal with 50% CP per t DM (FAOSTAT, 2013) was proportionately calculated for the substitutable amount in CLCA approach.

^c Ukrainian barley as marginal feed (Muñoz *et al.*, 2014; Schmidt and Brandao, 2013). Feed energy value and the equivalent mass were calculated as 15.2 and 11.9 MJ per kg DM for barley and alfalfa respectively (Møller *et al.*, 2005).

^d Marginal electricity = Coal as fuel type (Lund *et al.*, 2010; Mathiesen *et al.*, 2009).

^e Substituting marginal synthetic fertilizers: Calcium Ammonium Nitrate (CAN, Triple super phosphate (P₂O₅), Potassium Chloride (K₂O) (Hamelin *et al.*, 2012; Hamelin *et al.*, 2011; Tonini *et al.*, 2012).

^f Similar process flow as mentioned for avoided product, but consequential and allocation unit process data were used. See footnote 'a'.

^g Ethanol (99.7%) from biomass fermentation (Europe without Switzerland) adapted from Ecoinvent (Weidema *et al.*, 2013).

3

4

1 **Table 2.** Basic assumptions on the parameters considered in the inventory analysis.

| Parameters | Values | References |
|---|--|--|
| A. Lower heating value | | |
| - Bioethanol (MJ/kg) | 28.09 | (Cherubini and Ulgiati, 2010) |
| - Lignin (MJ/kg) | 22.9 | (Cherubini and Ulgiati, 2010) |
| - Methane (CH ₄) (MJ/m ³) | 35.8 | (Jørgensen, 2009) |
| - Lactic acid (assumed similar to organic acids) | 13 | (European Commission, 1990) |
| B. Parameters for biogas production: | | |
| i. C ₅ molasses (System A) | | (Drosg <i>et al.</i> , 2012) |
| - Total solids (TS) ^a | 31.1% | |
| - Volatile solids (VS) ^a | 30.1 % | |
| ii. Stillage fractions (System A) | | |
| - TS ^b | 12% | See footnote |
| - VS ^b | 10.2% | |
| iii. Residues from decanted press juice (System B) VS ^c | | 82% of DM ^c |
| C. Emission factors (g per MJ bioethanol production)^d | | |
| | NO _x = 38, CH ₄ = 1.5, N ₂ O = 0.8 | (Danish Energy Agency, 2012) |
| D. Heat and electricity input^e | | |
| i. Biogas digester | | Heat (H) = 1110 MJ _h Electricity (E) = 660 MJ _e (Berglund and Börjesson, 2006; Pugesgaard <i>et al.</i> , 2013) |
| ii. Combustion of lignin | | H = 40 MJ _h and E = 660 MJ _e Assumed from straw fired in CHP (Nielsen, 2004) |
| E. Nutrient content in the digestate, in g/kg digestate (System A and B)^f (N, P, K) | | |
| | 5, 0.9, 2.8 respectively | (Drosg <i>et al.</i> , 2015; O’Keeffe <i>et al.</i> , 2011) |
| F. Prices for computing allocation factors | | |
| - Bioethanol (Euro/MJ) ^g | 0.03 | |
| - Electricity (Euro /kWh) ^h | 0.25 | |
| - Heat (Euro/MJ) ⁱ | 0.03 | |
| - KCl (Euro/kg) ^j | 0.28 | |
| - Lactic acid (Euro/kg) ^k | 1.36 | |

| | |
|---------------------------------------|------|
| - Feed protein (Euro/kg) ^l | 0.33 |
| - Fodder silage (EUR/kg) ^m | 0.02 |

Assumptions:

^a TS and VS of the C₅ molasses are based on the total weight of molasses.

^b TS and VS of the stillage fractions are based on the total weight of stillage.

^c DM represents the substrate available for biogas after the decanted press juice (O'Keeffe et al., 2011) (SI-6, Fig. S-6.1)

^d Assumed similar to coal.

^e Energy input per t DM fuel.

^f NPK content (digestate) are per t fresh substrates (Fig. 1 and Fig. 2).

^g Average price of denaturated fuel ethanol for the period May 2006-Apr 2016 (index mundi, 2016; PURE) and validated with EUBIA (2016).

^h Price of electricity applied representative for the average Danish electricity price, including VAT and other recoverable taxes and levies the period of 2011-2015 (European Commission, 2012).

ⁱ Based on annual heat price of Denmark (Energitilsynet, 2012).

^j Average price of KCl, calculated for K using K to KCl molar ratio (May 2006-Apr 2016) (index mundi, 2016).

^k Price taken after Refs. (Lynd *et al.*, 2005; Wee *et al.*, 2006). Lactic acid considered a purity level of 90% (Kamm *et al.*, 2009).

^l Price of protein based on soybean meal (May 2006-Apr 2016) (index mundi, 2016; Statistics Denmark, 2016). Danish database represents feed compound for cattle (except calves, with high protein content). Price proportionately calculated for the crude protein content of soymeal (50% of the DM) (Dalgaard *et al.*, 2007) and protein extracted from the GBR (65% CP of the DM of the protein cake) (O'Keeffe et al., 2011).

^m Silage fodder traded in Denmark from 2005-2011 (Statistics Denmark, 2016).

1 **Table 3.** Primary input and output of materials related to the conversion of 1 t straw (with 85% DM)
 2 to bioethanol (System A), all data are per 1 t straw.

| Materials | Units | Amount |
|---|-----------------------|--------------------------------------|
| A. Input | | |
| Straw | t (85% DM) | 1 |
| Water ^a | kg | 2747 |
| Enzyme ^a | kg | 40 |
| Energy^b | | |
| - Heat | MJ _h | 4071 |
| - Electricity (kWh elec/t straw) | MJ _e | 850 |
| Additives | | |
| | | kg |
| - Diaammonium Phosphate (DAP) ^c | | 1.87 |
| - Corn steep liquor ^c | | 14.2 |
| - NaOH (49%) ^b | | 0.53 |
| - Ammonia water (25%) ^b | | 1.76 |
| B. Output (Primary) | | |
| - Bioethanol ^a | kg | 186 |
| - C ₅ molasses + residues from stillage ^d | kg | 392 |
| - Lignin ^d | kg | 152 |
| - KCl ^e | | 12 |
| Emissions^d | | |
| - During biomass production | | |
| ○ N ₂ O-N (total) | kg CO ₂ eq | -15 ^f 67 ^g |
| ○ due to SOC change | kg CO ₂ eq | 143 ^f -59 ^g |
| ○ N-leaching (NO ₃ -N) | kg N | 1.4 ^g |
| ○ P losses | kg P | 0.06 ^g |
| - During biomass conversion | | |
| ○ CO ₂ | kg CO ₂ eq | 162 |
| ○ Ethanol | kg | 12 |
| - | | |

Assumptions:

^a Average of the studies from Bentsen *et al.* (2006), Kaparaju *et al.* (2009) and Wang *et al.* (2013).

^b Based on Bentsen *et al.* (2006).

^c Based on Wang *et al.* (2013).

^d Based on Bentsen *et al.* (2006) and Kaparaju *et al.* (2009).

^e Total mass from the hydrolysate and stillage fractions. Recovery rate 90% (Larsen *et al.*, 2008).

^f Emissions are related to consequences of straw removal (Parajuli *et al.*, 2016). Negative value indicates mitigation potential. Total N₂O-N emission (direct + indirect) = -0.033 kg N₂O-N t DM⁻¹.

^g Emissions represented for ALCA approach. Emissions calculated for winter wheat were economically allocated to straw, i.e. 19% of winter wheat (Parajuli *et al.*, 2016). Negative value indicates soil C sequestration. Other related emissions per t DM: N₂O-N emission (direct + indirect) = 0.22 kg N₂O-N; NH₃-emission = 0.031 kg NH₃-N; NO_x-N = 0.1107 kg (Parajuli *et al.*, 2016). Detailed on the other material inputs are shown in SI-1, Table S-1.1.

1

2

1 **Table 4.** Input-output of materials for the conversion of 1 t DM alfalfa to biobased lactic acid (system
 2 B), all data are per t DM of alfalfa.

| Materials | Unit | Amount | Remarks |
|-----------------------------------|-----------------------|-------------------|---|
| Input | | | |
| Alfalfa | t DM | 1 | |
| Energy | | | |
| - Heat, steam ^a | MJ _h | 126 | |
| - Electricity ^a | MJ _e | 211 | |
| Fermentation media ^b | kg | 5.94 | |
| Enzyme | kg | 18 | |
| Water ^c | kg | 450 | |
| Output^d | | | |
| Lactic acid | kg DM | 89 | 99 kg (DM 90%) |
| Feed protein | kg DM | 26 | DM 40%, fodder protein the pure form of CP (65%). |
| Fodder silage | kg DM | 261 | 652 kg (DM 40%) |
| VS, residues for biogas | kg DM | 152 | Fresh weight 2.95 t (6% DM) |
| Emissions | | | |
| - During biomass production | | | |
| - Due to SOC change | kg CO ₂ eq | - 37 ^e | (Parajuli et al., 2016) |
| - N ₂ O-N | kg CO ₂ eq | 13 | (Parajuli et al., 2016) |
| - N-leaching (NO ₃ -N) | kg N | 3.4 | (Parajuli et al., 2016) |
| - P losses | kg P | 0.13 | (Parajuli et al., 2016) |

Assumptions:

^a Calculated based on O'Keeffe et al. (2011) and Kamm et al. (2009).

^b Calculated based on Kamm *et al.* (2010) (see section 2.5.1).

^c Calculated also considering the re-circulated water (O'Keeffe et al., 2011).

^d Products output calculated based on O'Keeffe et al. (2011) and Kamm et al. (2009) for 1 t DM of alfalfa (with 35% DM at harvest). Feed protein, estimated equivalent to soymeal (based on CP content) (Table 2). = 34 kg DM.

^e Calculated from Parajuli et al. (2016); negative value indicates soil C sequestration. Emissions per t DM : N₂O-N= 0.03 kg; NH₃-N = 0.04 kg; NO_x-N = 0.01 kg (Parajuli et al., 2016).

1 **Table 5:** Energy balance calculated for the biorefinery plants. The balance accounted all useful
 2 energy consumption within the biorefinery systems.

| | Units | Amount ₅ |
|---|-----------------|---------------------|
| A. System A | | |
| Total energy input (per 1 t, 85% DM straw) | | |
| - Heat | GJ _h | 4.1 |
| - Electricity | Gj _e | 0.8 |
| Total energy output | | |
| - Heat | GJ _h | 2.1 |
| - Electricity | Gj _e | 1.7 |
| Deficit/surplus | | |
| - Heat | GJ _h | -2.01 |
| - Electricity | Gj _e | 0.83 |
| B. System B | | |
| Total Energy input (per 1 t DM, alfalfa) | | |
| - Heat | GJ _h | 0.86 |
| - Electricity | Gj _e | 0.31 |
| Total energy output | | |
| - Heat | GJ _h | 0.59 |
| - Electricity | Gj _e | 0.84 |
| Deficit/surplus | | |
| - Heat | GJ _h | -0.99 |
| - Electricity | Gj _e | 0.53 |
| C. Net balance (System C) | | |
| - Heat | GJ _h | -2.78 |
| - Electricity | Gj _e | 1.3 |

4

5

1 **Table 6.** Environmental impact potentials of producing bioethanol and lactic acid from standalone plants and from system C (FU is “1 MJ_{EtOH} + 1 kg_{LA}”).
 2 Negative values indicate the environmental impact abatement potentials.

| Impact categories | Units | CLCA | | | ALCA | | |
|--------------------|-----------------------|--------------------------------------|---------------------------------------|--------------------------------------|----------------------|----------------------|--------------------|
| | | System A | System B | System C | System A | System B | System C |
| GWP ₁₀₀ | kg CO ₂ eq | | | | | | |
| - without iLUC | | 0.13 (0.16) ^a | 0.39 (1.81) ^a | 0.05 (0.15) ^a | 3.78 | 4.4 | 3.02 |
| - with iLUC | | - | 0.64 | 0.29 | 3.85 | 4.7 | 3.43 |
| | | 1.5*10 ⁻⁴ | -1.4 *10 ⁻³ | 1.3*10 ⁻⁵ | | | |
| EP | kg PO ₄ eq | (1.8*10 ⁻⁴) ^a | (3.7 *10 ⁻³) ^a | (2.9*10 ⁻⁴) ^a | 4.9*10 ⁻³ | 1.4*10 ⁻² | 5*10 ⁻³ |
| NRE use | MJ eq | 1.25 (1.51) ^a | 14.63 (29.22) ^a | 0.38 (1.39) ^a | 45 | 65 | 31 |
| ALO | m ² a | 0.02 (0.02) ^a | 1.99 (3.33) ^a | 0.11 (0.17) ^a | 2.68 | 8.6 | 5.31 |

^a Values in the parenthesis are the gross impacts, i.e. without avoided impacts.

3

4

1 **Table 7.** Contribution patterns of the co-products for avoiding and sharing the environmental
2 impacts within CLCA and ALCA approach. Units per FU in each system are: net GWP₁₀₀ = kg CO₂ eq,
3 EP = kg PO₄ eq, NRE use = MJ eq , and ALO = m².

| Contributions | CLCA | | | | ALCA | | | |
|--------------------|---------------------|---------------------|---------------------|---------------------|--------------------|---------------------|--------------------|--------------------|
| | Electricity | Recovered nutrients | Feed protein | Fodder silage | Electricity | Recovered nutrients | Feed protein | Fodder silage |
| System A | | | | | | | | |
| GWP ₁₀₀ | -0.03 | -0.001 | - | - | 0.68 | 0.81 | - | - |
| EP | -3*10 ⁻⁵ | -9*10 ⁻⁷ | - | - | 9*10 ⁻⁴ | 1*10 ⁻³ | - | - |
| NRE use | -0.25 | -0.02 | - | - | 8.18 | 9.69 | - | - |
| ALO | -2*10 ⁻⁵ | -5*10 ⁻⁸ | - | - | 0.48 | 0.57 | - | - |
| System B | | | | | | | | |
| GWP ₁₀₀ | -0.58 | -0.05 | -0.12 | -0.67 | 0.54 | 0.03 | 0.58 | 0.05 |
| EP | -8*10 ⁻⁴ | -6*10 ⁻⁵ | -9*10 ⁻⁴ | -3*10 ⁻³ | 2*10 ⁻³ | 9*10 ⁻⁵ | 2*10 ⁻³ | 2*10 ⁻⁴ |
| NRE use | -5.54 | -0.33 | -0.73 | -8 | 7.95 | 0.46 | 8.64 | 0.72 |
| ALO | -5*10 ⁻⁴ | -2*10 ⁻⁵ | -0.54 | -0.79 | 1.01 | 0.07 | 1.25 | 0.08 |
| System C | | | | | | | | |
| GWP ₁₀₀ | -0.06 | -0.013 | -0.01 | -0.03 | 0.55 | 0.66 | 0.71 | 0.04 |
| EP | -7*10 ⁻⁵ | -2*10 ⁻⁵ | -4*10 ⁻⁵ | -1*10 ⁻⁴ | 9*10 ⁻⁴ | 1*10 ⁻³ | 1*10 ⁻³ | 7*10 ⁻⁵ |
| NRE use | -0.54 | -0.1 | -0.03 | -0.34 | 5.66 | 6.79 | 7.25 | 0.45 |
| ALO | -5*10 ⁻⁵ | -5*10 ⁻⁵ | -0.02 | -0.04 | 0.96 | 1.17 | 1.25 | 0.08 |

1 **Table 8.** Results obtained from the sensitivity analysis. Units per FU of each system are: net GWP₁₀₀
 2 = kg CO₂ eq and NRE use = MJ eq.

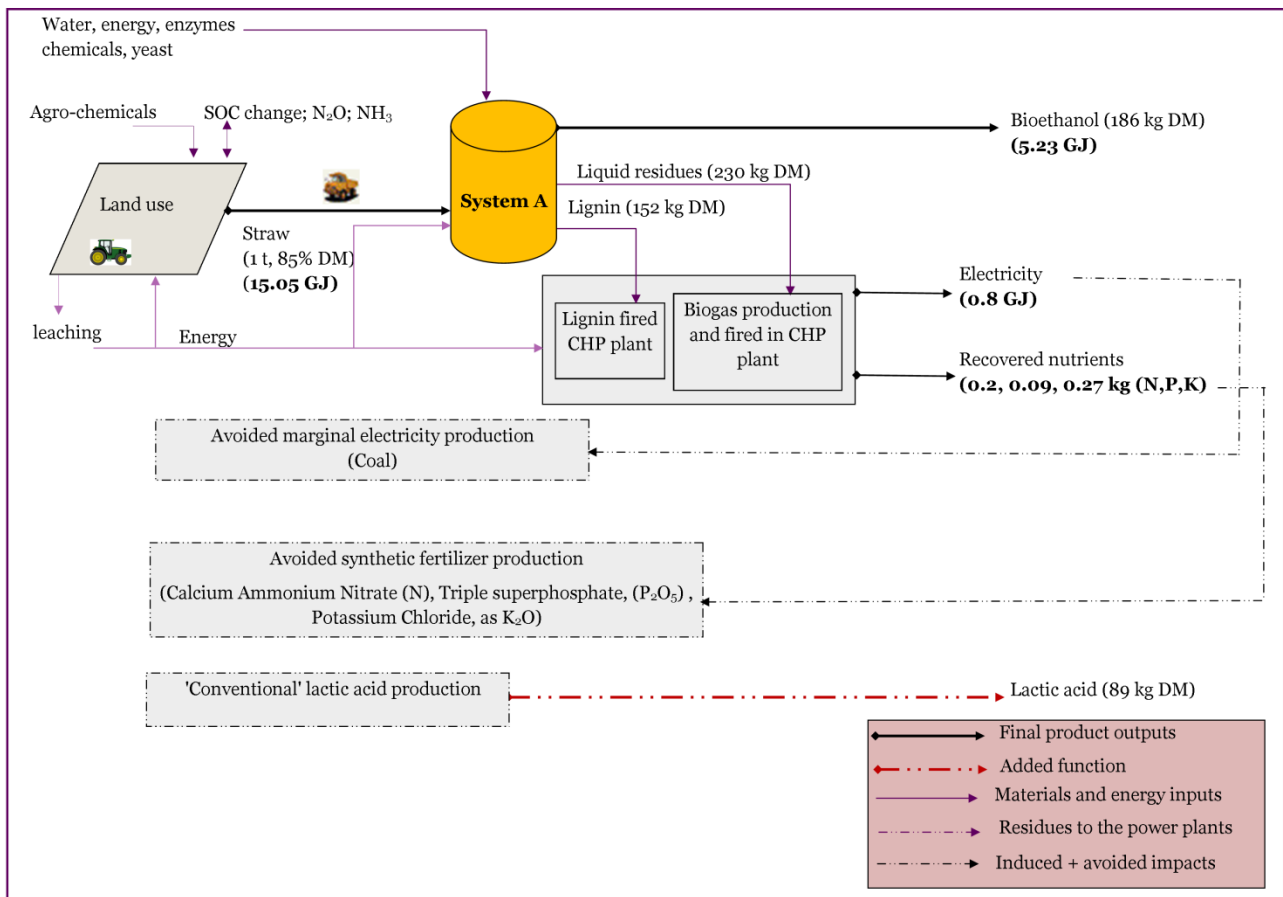
| | Basic Scenario | Marginal products | | Variations in the yield | | |
|--------------------------|-------------------|--------------------------|-------------------------------|-------------------------|------------|------------|
| | | Electricity ^a | Fodder silage ^b | +14 % EtOH | +10% LA | -10% LA |
| System A (per FU) | | | | | | |
| Net GWP ₁₀₀ | 0.13 | 0.14 | - | 0.11 | 0.12 | 0.14 |
| Net NRE use | 1.25 | 1.24 | - | 1.09 | 1.15 | 1.36 |
| System B (per FU) | | | | | | |
| Net GWP ₁₀₀ | 0.39 | 0.67 | 0.63 | 0.34 | 0.35 | 0.43 |
| Net NRE use | 14.63 | 14.43 | 17 | 12.8 | 13.3 | 16.3 |
| System C (per FU) | | | | | | |
| Net GWP ₁₀₀ | 0.05 | 0.08 | 0.06 | 0.05 | 0.04 | 0.05 |
| Net NRE use | 0.38 | 0.4 | 0.55 | 0.29 | 0.33 | 0.4 |

^a Natural gas as source for marginal electricity production

^b Grass-silage as a source of energy-feed.

3

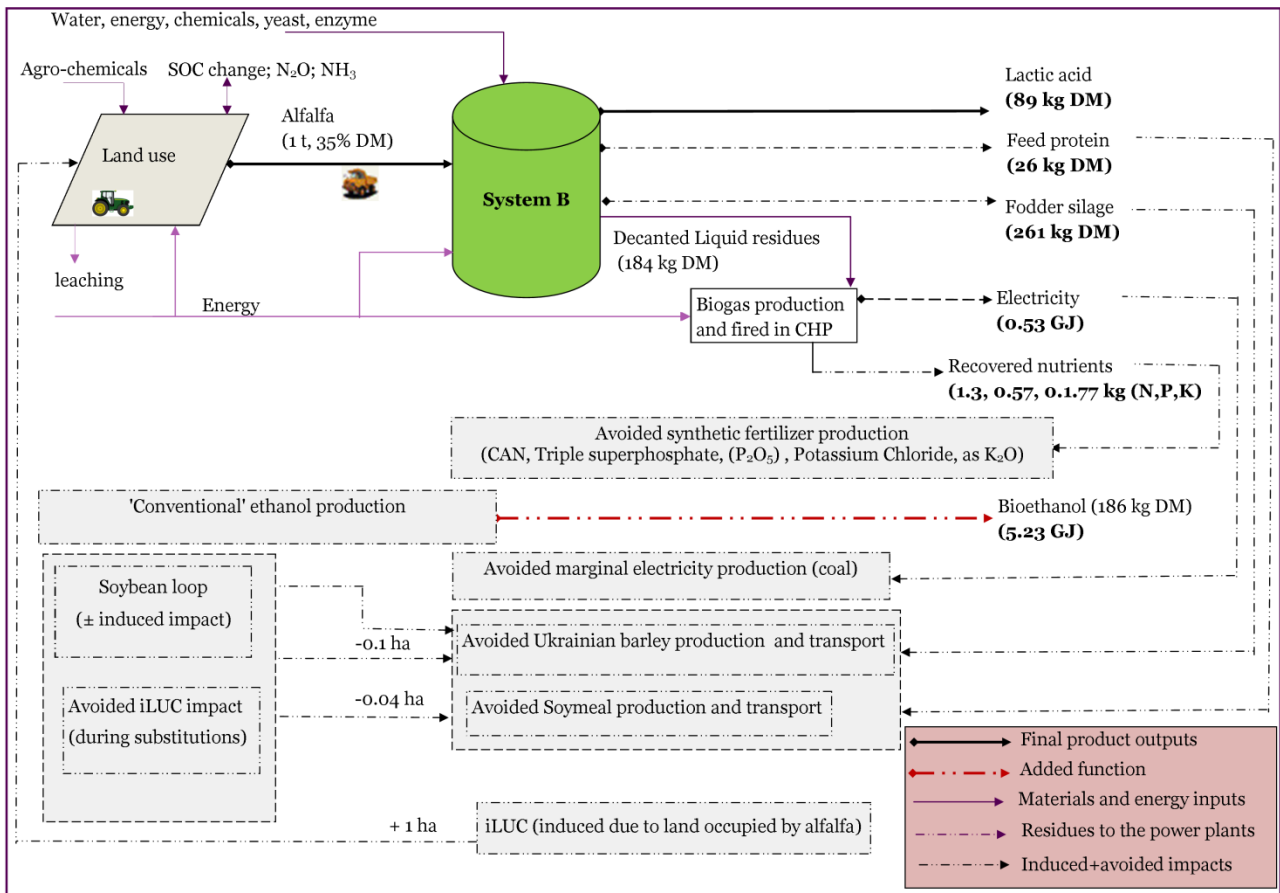
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2 **Fig. 1.**

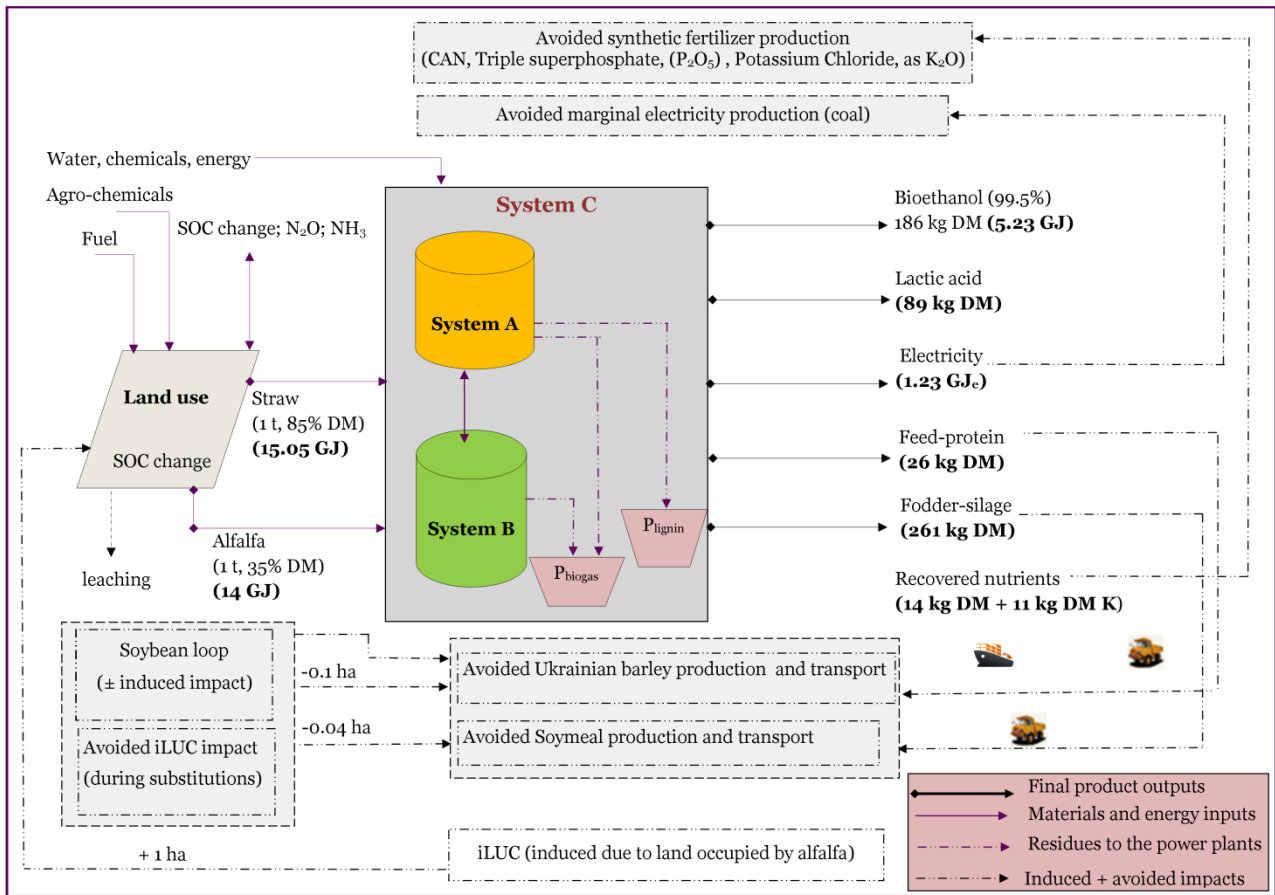
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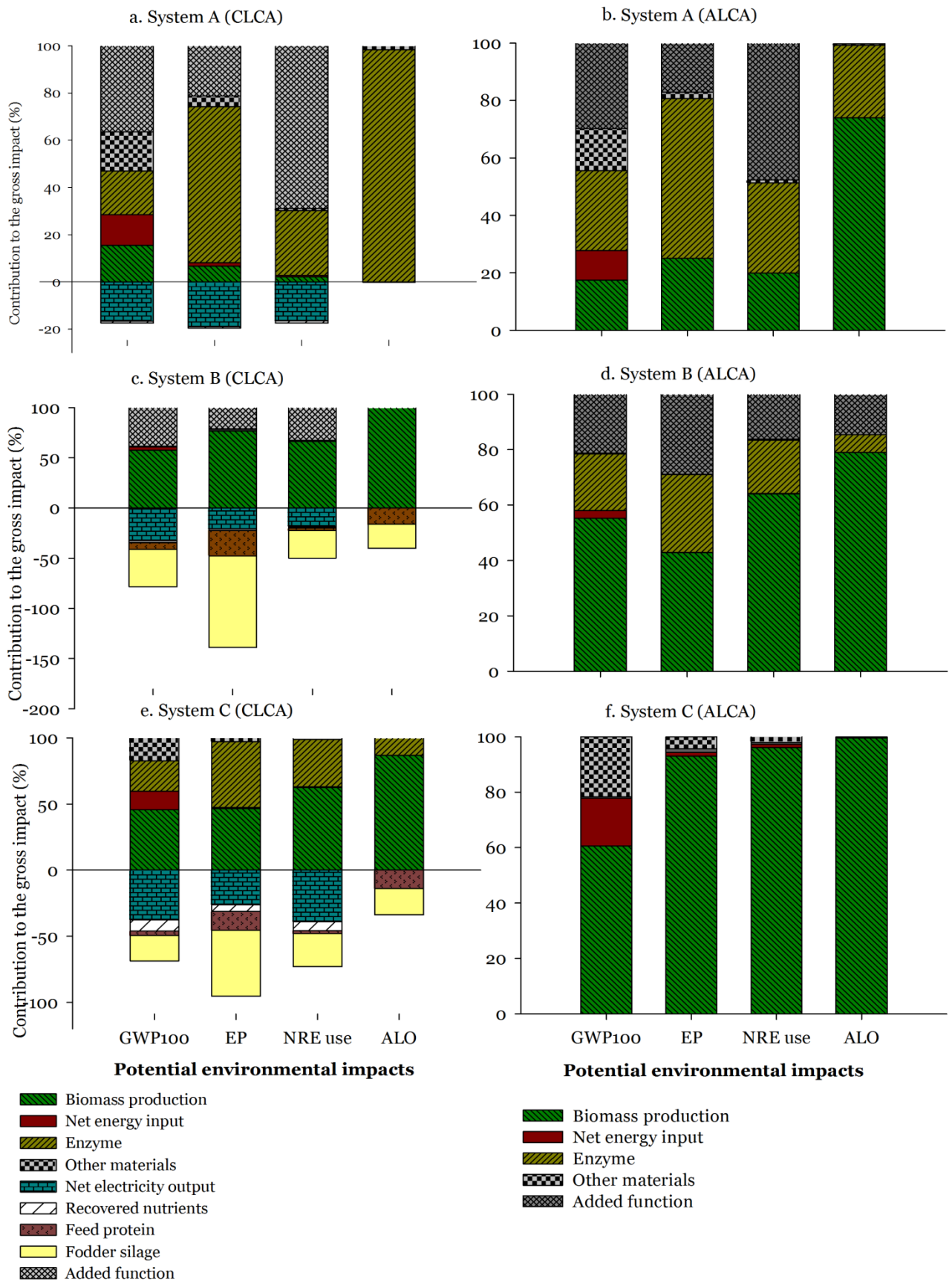
2 Fig. 2.

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Fig. 3.



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Fig. 4.

1 **Appendix A: Supplementary data (SI):**

2 Environmental impacts of producing bioethanol and biobased lactic acid from standalone
3 and integrated biorefineries using a consequential and an attributional life cycle assessment
4 approach

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1 **SI-1.** Data for biomass production

2 **Table S-1.1.** Input-output of the materials flow assumed for the alfalfa production, per 1 t

3 DM, summarised after Parajuli *et al.* (2017).

| | Units | Alfalfa | Comments/Remarks |
|-----------------------------------|-----------------------------|---------|--|
| Biomass output | t DM/ha | 12.02 | (Møller <i>et al.</i> , 2005; NaturErhvervstyrelsen, 2015) |
| Farm inputs | | | |
| Synthetic fertilizer ^a | kg/ t DM | | SEGES (2010) |
| N | | - | |
| P | | 3 | |
| K | | 18 | |
| Lime | kg/ t DM | 4.57 | Based on Hamelin <i>et al.</i> (2012) |
| Pesticides | kg/ t DM | 0.02 | Based on SEGES (2010) |
| Direct primary energy input | MJ/ t DM | 343 | Field preparation and harvesting |
| Transport materials | t km/ t DM | 6 | (seed +agri-chemicals) |
| Emissions | | | (Parajuli <i>et al.</i> , 2017) |
| N ₂ O | kg CO ₂ eq/ t DM | 16 | |
| SOC change | kg CO ₂ eq/ t DM | -37 | |
| Leaching | kg N/t DM | 3.4 | |
| P losses | kg P/t DM | 0.14 | |
| Transport biomass | t km/t DM | 200 | field to the biorefinery plant |

4

5

1 **SI-2. Parameters and calculation details on the methane yield from the residues**

2 System A: The volatile solids (VS) (%) in the stillage fractions of the total wet weight of the
3 stillage was based on Kaparaju *et al.* (2009) (Table 2). In the case of molasses, of the total
4 wet-weight of the cake the total solids (TS) and VS were assumed, as reported in Drogg *et al.*
5 (2012) (Table 2). The total nitrogen content in the stillage was assumed as 1.78 g per kg_{EtOH}
6 (Kaparaju *et al.*, 2009), hence ammonia inhibition would be 0.2 g per kg_{EtOH}. It is also
7 accelerated with higher temperature. Thus it is wise to avoid thermophilic process
8 temperatures when treating nitrous feedstock such as stillage, hence mesophilic reactors was
9 considered and the conversion efficiency was set lower as explained below.

10 System B: The total mass of fermentable substrate for the production of methane (CH₄) was
11 based on the VS (%) in the decanted press juice (O’Keeffe *et al.*, 2011) (see Table S-1.1).

12 System C: The substrate available for biogas conversion in System C is the combination of
13 residual resources available from System A and System B (Fig. S-5.1 and Fig. S-6.1).

14 The total potential methane yield was calculated by utilizing Eq. (i) (Pugesgaard *et al.*, 2013).

15
$$CH_4 (potential) = \varepsilon * VS * B_0 * 0.67 \dots\dots\dots Eq. (i)$$

16 where, CH₄ (potential) = methane production (kg); VS (in kg, see Table 2); B₀ is the
17 maximum methane-producing capacity of the added material (m³kgVS⁻¹) (see Table S-1.1); ε
18 = process efficiency = 0.8, based on the average efficiency of hydrolysate and stillage
19 fractions, as reported in Kaparaju *et al.* (2009) and 0.67 was the conversion factor from
20 volume to kg CH₄ (Olesen *et al.*, 2004). The energy input to biogas plant was based on
21 Berglund and Börjesson (2006). Methane loss during combustion was set to 1.8% of the total
22 conversion (Pugesgaard *et al.*, 2013). Conversion of biogas to heat and electricity was 18.69
23 MJ_h and 26.7 MJ_e respectively per kg of CH₄, with LHV of CH₄ set as 35.8 MJm⁻³, and the
24 heat and electricity conversion efficiency were set to 35% and 50% (Jørgensen, 2009).
25 Likewise, the amount of substrate available from the GBR to the biogas digester was
26 calculated based on the studies of O’Keeffe *et al.* (2011), Kamm *et al.* (2009) and (Kamm *et*
27 *al.*, 2010) (Fig. S-6.1). In this case, the B₀ of the added material was assumed 39.5%,
28 estimated based on Pugesgaard *et al.* (2013) for crop residues

29

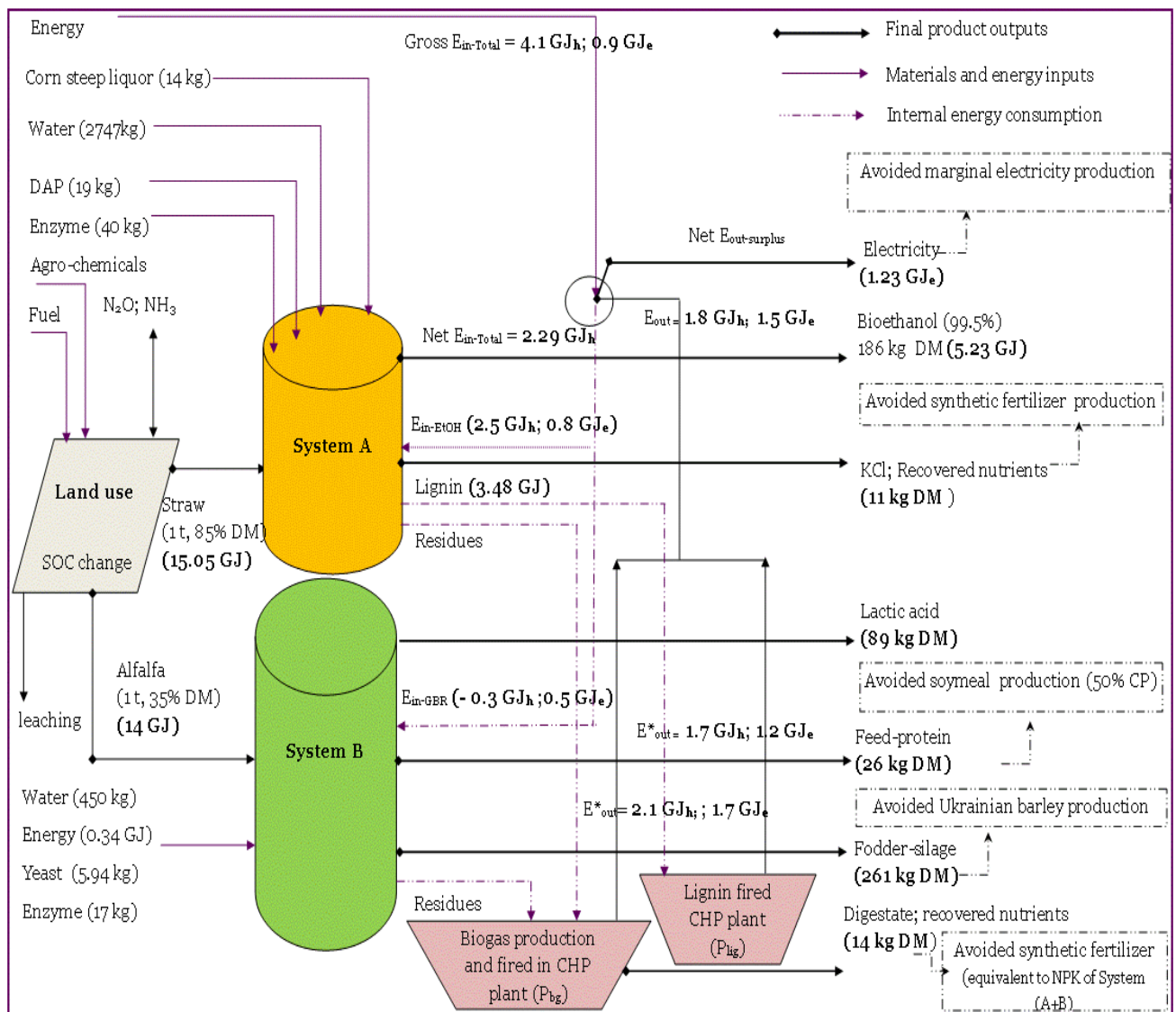
- 1 **SI-3.** Supporting parameters and energetic inputs considered in the basic scenario
 2 **Table S-3.1:** Energy inputs during the processing of biomass in System B; calculated based
 3 on O’Keeffe et al. (2011) and Kamm et al. (2009).

| Biomass Processing and stages in GBR | Units | per t DM alfalfa |
|--------------------------------------|-------|------------------|
| Pumping | kWh/t | 0.69 |
| Fiber processing to silage fodder | | |
| Pressing | kWh/t | 4.9 |
| Protein extraction | | |
| Steam coagulation | MJ/t | 126 |
| Skimming | kWh/t | 1.31 |
| Centrifuging | kWh/t | 3.41 |
| Decanting | kWh/t | 1.03 |
| Lactic acid production | | |
| Stirring | kWh/t | 3.75 |
| Ultrafiltration | kWh/t | 4.85 |
| Bipolar electrodialysis | kWh/t | 33 |
| Reverse osmosis | kWh/t | 4.28 |
| Distillation | kWh/t | 1.32 |

4

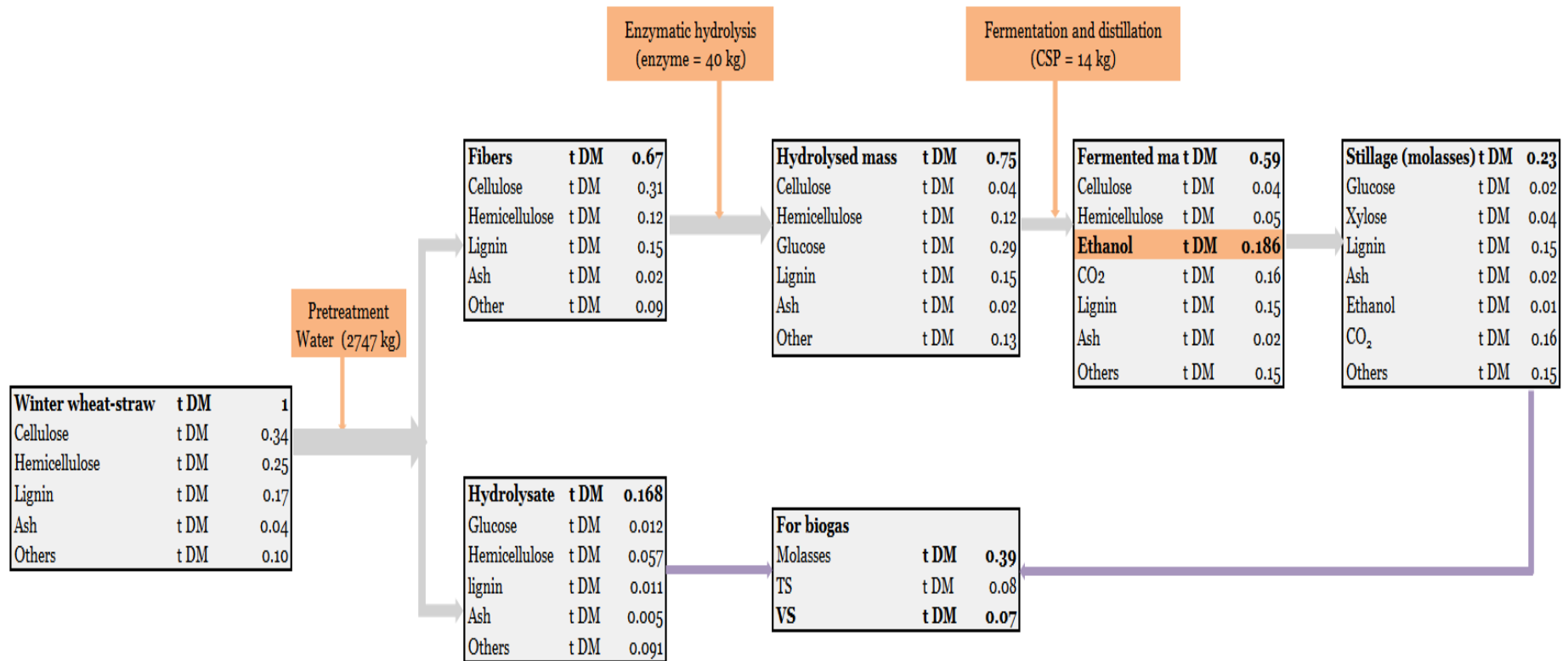
5

1 **SI-4. Process flow diagram of the standalone systems and the integrated system**
 2 The details on the energy flows within the biorefinery systems and the energy exchanges
 3 between the two standalone systems for System C are shown in Fig. S-4.1. The notations as
 4 mentioned in the Fig. S-4.1 are: Gross $E_{in-Total}$ = total energy required in the biorefinery
 5 systems; E_{out} = Energy produced from the CHP plants (after deducting the self-demand, e.g.
 6 to burn the fuel); E_{in-GBR} = Energy input to System B; $E_{in-EtOH}$ = Energy input to System A;
 7 E^*_{out} = co-produced energy from the CHP plants; Net $E_{in-Total}$ = Energy required in the
 8 biorefinery after accounting all internal consumptions and Net $E_{out-surplus}$ = surplus electricity
 9 production from the system



10
 11 **Fig. S-4.1. Materials and energy flows in the integrated system (System C). Electricity**
 12 **produced is the net values after utilizing the internal consumptions in each standalone**
 13 **systems.**

1 **SI-5. Transformation of biomass in System A**



2

3 **Fig. S-5.1.** Mass flow for the conversion of straw to bioethanol, mass balance averaged from studies Bentsen *et al.* (2006), Kaparaju *et al.*
4 (2009) and Wang *et al.* (2013).
5

1 **SI-6.** Transformation of biomass in System B

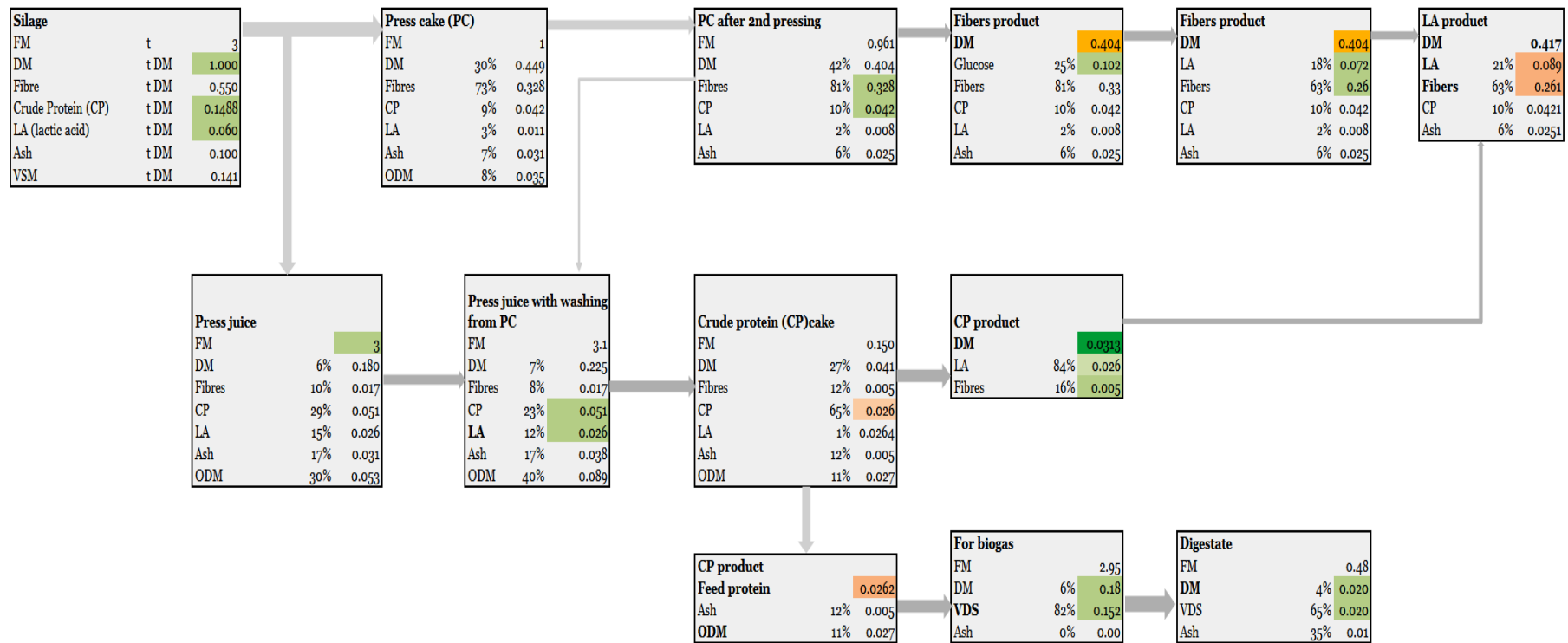
2 Regarding the processes involved for the production of lactic acid, it was assumed that it
3 initiates with handling of alfalfa with mechanical processing, including the chopping of the
4 biomass. The process was assumed to be followed by the extraction of press-juice (DM 5 %) and
5 press-cake from a mechanical screw-press (O’Keeffe et al., 2011). The fractions of press
6 juice and the press cake were assumed to be 70% and 30% of the fresh matter respectively
7 (O’Keeffe et al., 2011). Fiber losses (5% of the total fibers in biomass) during the washing
8 steps were assumed to be recovered and utilized as residues for biogas production (see Fig. S-
9 6.1). Press juice-stream was divided into two sub-streams, one for the protein extraction and
10 another for the lactic acid production (Kamm et al., 2009; O’Keeffe et al., 2011). After the
11 separation of press-juice and press cake, the DM content in the press cake (i.e. after the 2nd
12 pressing) was assumed for hydrothermal pre-treatment process, followed by enzymatic
13 hydrolysis. The amount of enzyme used during the hydrolysis process was calculated based
14 on the cellulose content of the press cake. Enzyme loading per cellulose content of the
15 pretreated biomass (Bentsen et al., 2006; Kaparaju et al., 2009) was considered to calculate
16 the amount of enzyme. The calculated mass of enzyme was 17 kg per ton dry biomass, and
17 was comparable to the loading rate of 20 kg per ton biomass, as reported in Wolfrum *et al.*
18 (2013). It was also close to the value reported (51 kg per kg press cake), reported in Duque
19 (2016). Cellulose content in the press cake was assumed 33% of the total fibers (DM) of the
20 press cake (PC) (see Fig S-6.1). The assumption was within the range (20-36%), as reported
21 in Xiu *et al.* (2014). It was reported to be around 44% in clover grass cake (reported for the
22 initial hour of pretreatment) (Duque, 2016). The sugar content in the fibers (input and output
23 streams) (Table S-6.1) was assumed after Cybulska *et al.* (2010). Considering these, glucose
24 in the liquid and solid fractions (i.e. of the hydrolysed materials) were assumed 1.3% and 32%
25 respectively of the sugar content in the press cake (Table S-6.1). Considering the uncertainty
26 to harness glucose in both fractions, in the current study only the glucose contained in the
27 solid fractions was considered for the fermentation process. Yield of glucose after the
28 hydrolysis process was estimated to be 125 kg per t DM of the solid fraction (see Fig. S-6.1
29 and Table S-6.1). Regarding the fermentation of the substrate, a lactic acid producing

1 bacteria can be considered (Pahlow *et al.*, 2003). In order to overcome the inhibitory effect
 2 during the fermentation process, fermentation takes can be carried out in a continuous
 3 dialysis process or in an electro-dialysis system (Kim and Moon, 2001). In the current study,
 4 for the recovery of lactic acid and protein following processes were considered: ultrafiltration,
 5 reverse osmosis (Patel *et al.*, 2006), bipolar electro-dialysis and distillation (Kim and Moon,
 6 2001). Energy consumption for these processes is summarized in Table S-3.1. Feed protein
 7 contained in the fermentation broth was assumed to be separated using an ultrafiltration
 8 membrane technology (Li *et al.*, 2006). Sodium hydroxide was used as a base material, which
 9 results into sodium lactate. The total yield of lactic acid was calculated from the press-juice
 10 fraction and the fraction obtained after the fermentation of glucose, as stated above. Crude
 11 lactic acid (26 kg lactic acid per t DM) contained in the press juice with an extraction
 12 efficiency of 70% resulted to produce 18 kg LA (DM) (O’Keeffe *et al.*, 2011). Likewise,
 13 considering the glucose to lactic acid conversion factor to be 79% (Doran-Peterson *et al.*,
 14 2008), the total production of biobased lactic acid was estimated to be 89 kg (SI-6). Liquid
 15 residue produced from the processing of biomass was considered as substrate for biogas
 16 production.

17 **Table S-6.1:** Material balance after the conversion of dry matter of the press cake
 18 undergoing enzymatic hydrolysis process, calculation based on Cybulska *et al.* (2010).

| Components | Input ^a | | Output | | | |
|---------------|--------------------|-----|--------|-------|-------|-----|
| | | | Liquid | | Solid | |
| | % | kg | % | kg | % | kg |
| Glucose | 33% | 133 | 1.3% | 5 | 31% | 125 |
| Hemicellulose | 15.60% | 63 | 5.90% | 24 | 9.70% | 39 |
| Lignin | 21% | 85 | 18.50% | 75 | 2.50% | 10 |
| Ash | 5.65% | 23 | 0.00% | 0.000 | 5.65% | 23 |

19



1

2 **Fig.S-6.1:** Mass flow for the conversion of alfalfa to lactic acid and other biobased products, data partly adapted from (O’Keeffe et al., 2011) and
 3 adjusted for DM content of fresh alfalfa to 35%. Glucose content were based on Cybulska et al. (2010) (see text and Table S-6.1), and the
 4 conversion factor of the glucose in the associated fraction was based on Doran-Peterson et al. (2008). The production of LA as presented in the
 5 fraction “press juice with washing from press cake (PC)” indicates the yield assumed after enzymatic hydrolysis plus the conversion of crude LA
 6 contained in the press juice. ‘Green’ shaded parts represent the depending intermediate particles and ‘orange’ shaded parts indicate the final
 7 product. All data presented are in t DM, calculated from the fresh matter (FM), composition of each chemicals are with respect to the DM
 8 mentioned.

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