Technical, economic and environmental assessement of bioethanol biorefinery from waste biomass

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Highlights

- The sustainability of bioethanol biorefinery from waste biomass was assessed
- The technical evaluation lead to yields equal to 0.14-0.22 kg bioethanol/kg biomass
- Economic sustainability was accomplished for all waste biomasses considered
- Life Cycle Analysis showed that manure achieved the best environmental performances
- Sugarcane achieved the lowest energy input consumption (64 %)

TECHNICAL, ECONOMIC AND ENVIRONMENTAL ASSESSEMENT OF BIOETHANOL BIOREFINERY FROM WASTE BIOMASS

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Abstract

This study presents a sequential three-steps methodology for the technical, economic and environmental assessment (TEEA) of bioethanol production from waste biomass. In EU the most abundant waste biomasses produced in 2018 could be ascribed to three main categories: lignocellulosic (329.41 Mt), starch (160 Mt) and sugar-based (58.56 Mt). The technical assessment compiled an inventory of the waste biomasses and subsequently designed their biological conversion into ethanol through integrated biorefinery processes by means of material flow analysis (MFA); the economic assessment was aimed at the definition of the cut-off size of the biorefinery plant necessary to achieve profitability; the environmental assessment was based on Life Cycle Analysis (LCA) and energy balance (i.e. energy input consumption). For each of the three waste biomass categories, at least one that was significant as available amount and representative in terms of physico-chemical characteristics, was evaluated: sugarcane for sugar-based, potatoes for starch-based and rice straw, cattle manure and organic fraction of municipal solid waste (OFMSW) for lignocellulosic biomasses. The technical assessment of the biorefinery routes lead to the following yields (kg of bioethanol per kg of biomass): 0.16, 0.17, 0.22, 0.19 and 0.14 respectively. The economic profitability was reached by all biorefineries and Net Present Value (M€) were: 0.85 for sugarcane, 0.11 for potatoes, 0.09 for rice straw, 0.11 for cattle manure and 0.39 for OFMSW. From the environmental perspective, cattle manure reached the highest reduction of climate change and acidification impacts compared to other biomasses, while sugarcane achieved the lowest energy input consumption (around 64 %).

Keywords: biorefinery; economic analysis; energy balance; life cycle analysis (LCA); material flow analysis (MFA).

1. Introduction

Bioethanol industrial production is well-known by chemical engineers and its development faced alternate fortune. After a huge growth in the USA at the beginning of the XIX century, bioethanol production was marginalised by the taxes imposed by Civil War, later reinforced in the 1970s during the oil crisis and it recently bounced back. The USA Energy Information Administration stated that from 2006 to 2018 the consumption and production of bioethanol significantly increased (+60 % and +55 % respectively) (EIA, 2018), with a market value equal to 0.91 €/kg for 99.7 % v/v purity (PubChem, 2018). Leading bioethanol producers are USA (40 Mm^3 from corn and wheat) and Brazil

(25 Mm^3 from sugarcane) (Roy and Dutta, 2019). Bioethanol can be used as fuel in vehicles without any modification (Thangavelu et al., 2016) if blended with gasoline (from 3 % up to 20 %) (Roy and Dutta, 2019), increasing the octane number and avoiding the addition of methyl tertbutyl ether, thus fostering a cleaner combustion (European Biomass Industry Association, 2018). Biofuels represent a promising alternative to fossil fuels (Aditiya et al., 2016), being based on renewable feedstock and thus allowing a reduction of greenhouse gas (GHG) emissions. However, the production of biofuels from food and energy crops could be responsible for food insecurity and their cost is the main barrier to their promotion (e.g. in January 2020 gasoline was sold in the USA at 2.59 USD/gallon, ethanol E85 at 2.28 and biodiesel B20 at 2.89) (USA Department of Energy, 2020). In this framework, the development of biofuels production technologies from waste biomass, particularly within integrated biorefinery systems, could be extremely interesting.

Bioethanol production is based on three main biomass categories: sugar (sugarcane, beetroot, molasse) (Balat, 2011; Zabed et al., 2017), starch (corn, wheat, cassava) (Mohapatra et al., 2017) and lignocellulosic (energy crops, agricultural waste, organic fraction of municipal waste OFMSW) (Hafid et al., 2017; Stichnothe and Azapagic, 2009; Zhao et al., 2018). Algae were also described as promising feedstock for bioethanol production (Jambo et al., 2016; Sirajunnisa and Surendhiran, 2016).

The environmental performances of bioethanol blended with gasoline could significantly contribute to lower CO_2 footprint of the transport sector (Phwan et al., 2018), however feedstock production may exhibit significant impacts (Zucaro et al., 2016). Therefore, a careful analysis of the perspectives of bioethanol as biofuel should include the specific assessment of its production. There is general agreement on the improvement of the environmental performances of bioethanol production if the biorefinery process includes the valorisation of coproducts and residues (Chang et al., 2017; Zhao et al., 2020). A life cycle analysis (LCA) of seven different technologies for producing bioethanol from corn stover showed that all configurations allowed a net saving of the global warming potential (GWP) (up to 900-1200 kg CO_2 eq/t dry corn stover) (Zhao et al., 2018). A review focused on LCA of bioethanol production from food crops (Roy and Dutta, 2019) revealed that most LCA studies concluded that biofuels significantly reduced GHG emissions if they replace fossil fuels, however they failed to account the impact of land use and specific GHG emissions related to biofuels. The same review pointed out that economic sustainability depends on the biorefinery plant size and on the efficient recovery of coproducts. Another review study concerning LCA of bioethanol production from lignocellulosic biomass (Wiloso et al., 2012) emphasized that while bioethanol usually performs better than fossil fuels about GHG emissions, LCA approaches were often controversial in considering upstream chain of biomass and choice of allocation methods. Also the choice of the functional unit (FU) in LCA could lead to very different outcomes; while most studies adopt 1 ton of feedstock or 1 ton of bioethanol as FU (and the compared scenarios usually involve other feedstocks and/or their management perspectives in case of waste biomass), if MJ of fuel equivalent is chosen as FU, the comparison involves fossil fuels on an equivalent energy basis. This means that high biogenic carbon content makes the bioethanol production system a carbon sequester (Stichnothe and Azapagic, 2009).

The social, environmental and economic impact of bioethanol production in China was recently investigated (Wang et al., 2020a and b), considering employment creation, economic stimulus and energy use; while 1G bioethanol created higher economic growth and more jobs, 2G bioethanol was associated to lower energy consumption (Wang et al., 2020a) and to lower water demand and land use and environmental impacts (Wang et al., 2020b). The LCA of bioethanol production from sugarcane and cassava in Thailand (Papong et al., 2017) showed that GHG emissions for bioethanol were around 26-39 kg CO_2 eq/GJ heat content of the fuel, and social advantages were also observed (+15-18 % total employment, +30-45% income generated associated to agricultural stage). The LCA of bioethanol production from the whole cassava plant through integrated biorefinery in China gave back competitive energy balance and good reproducibility and environmental performances (Lyu et al., 2020). The LCA of bioethanol co-production with succinic acid from lignocellulosic biomass in Greece found GHG emissions around 32 g CO_2 eq/MJ (Chrysikou et al., 2018). The LCA of bioethanol production from cattle manure in Brazil outlined that the use of waste biomass as feedstock of the biorefinery counterbalanced the environmental impacts of bioethanol production (de Azevedo et al., 2017). The sustainability assessment of bioethanol production from cassava and molasses in Thailand (Haputta et al., 2020) showed that increasing bioethanol production and use could create significant benefits to society (job creation and mitigation of environmental costs) and increase the gross domestic product, and a net benefit to society equivalent to nearly 2 billion USD in 2016-2026 was estimated. Considering a wider viewpoint, a recent review (Sharma et al., 2020) concerning the overall assessment of bioethanol 2G biorefinery from lignocellulosic biomass demonstrated several main important issues: 2G bioethanol biorefineries, although not reaching 3 % of its production, exhibit higher GHG reduction potential than 1G biorefineries; however the main bottlenecks are represented by the feedstock (e.g. availability, transport costs, quality consistency) and by the conversion technology (i.e. pre-treatment, fermentation, hydrolysis, purification), which often presents serious technical and economic challenges that need to be overcome and “tailor-made” on the specific feedstock.

The aim of this study was the design of a methodology for the evaluation of bioethanol production from different waste biomasses. The approach consisted in a sequential technical, economic and environmental assessment (TEEA) of bioethanol production. The considered catchment area was EU-28 in year 2016. To our knowledge, most literature studies considered individual viewpoints in bioethanol biorefinery (e.g. LCA or economic and/or social assessment in specific geographical contexts) achieving results that could be sometimes controversial or partially representative. Therefore, the novelty of the present study was in the methodology itself, which was the development of a TEEA as sequential assessment tool in terms of: 1) technical feasibility, 2) economic profitability at full-scale and 3) environmental sustainability not related to a specific geographical context but to the whole EU. The pillars of the proposed methodology were based on the main findings of literature review: specific interest for 2G bioethanol integrated biorefineries, having better environmental performances compared to 1G biorefineries (higher GHG reduction, lower energy demand); careful attention for feedstock selection, to achieve availability and consistency, and for the technical and economic feasibility not only of the conversion technology, but of the whole integrated biorefinery, from feedstock supply to waste flows management. The technical assessment had the goal to identify, among the most abundant waste biomasses available in EU-28 and based on sugar, starch and lignocellulose, the ones appropriate for the conversion into bioethanol through integrated biorefinery processes. Afterwards, the economic analysis was aimed at screening the biorefineries verified as technically feasible, to further define the cut-off plant size at full-scale to reach profitability according to the fed biomass. Finally, the environmental impacts were quantified through the LCA of the technically feasible and economically profitable biorefineries and the assessment of energy input consumption. The presented TEEA methodology was therefore made of three consequent phases, the results of each one defining the development of the next phase. In details, the first phase was the selection of specific waste biomasses through the investigation of the technical feasibility of bioethanol production through Material Flow Analysis (MFA); the second phase (i.e. the economic analysis) was specifically aimed at screening the biorefineries verified as technically feasible in phase 1 through MFA, to define the cut-off plant size at full-scale to reach profitability according to the fed biomass; the third phase, considering only the technically feasible and economically profitable biorefineries, was the environmental assessment (through LCA and the analysis of the energy input consumption). In conclusion, specifically considering the second phase of the methodology, a Life Cycle Cost Analysis would have given a different output, referred to the functional units chosen for the LCA (1 ton of biomass and 1000 L of bioethanol), while the goal of the here presented methodology was instead to define the minimum biorefinery plant size related to each selected biomass and necessary to achieve the economic profitability. Moreover, specifically considering the third phase of the methodology, the choice of two functional units aimed to prove the consistency of the results of the LCA performed from two complementary perspectives (e.g. the valorization of waste biomass and the production of bioethanol), considering that according to literature sometimes the results of LCA studies were controversial.

2. MATERIALS AND METHODS

2.1. Technical assessment

Three biomass categories were analysed: sugar, starch and lignocellulosic. The technical assessment consisted of two phases: a) definition of biomass inventory and b) material flow analysis (MFA) of biomass conversion into bioethanol. Firstly, an inventory of the available waste biomasses in EU-28, considering quantitative (Mt/y) and qualitative (physico-chemical) characteristics, was carried out. The quantitative part of the inventory was based on Eurostat and FAO databases and EU commission reports, while the qualitative analysis was gathered from the available scientific literature. The outcome of the first phase of the technical assessment was the selection of the most significant and representative biomasses (in terms of amount and composition) within each of the three categories (sugar, starch and lignocellulosic).

Secondly, to achieve consistency, bioethanol production was considered under the same boundary conditions through the biological conversion of the waste biomasses according to their physico-chemical composition. The bioethanol conversion process was defined according to 1) physico-chemical composition of the selected biomasses and 2) conversion process available in the scientific literature. The biorefinery conversion route consisted in different outlines depending on the type of biomass: fermentation and downstream processes for sugar biomass; hydrolysis, fermentation and downstream processes for starch biomass; pre-treatments, hydrolysis, fermentation and downstream processes for lignocellulosic biomass. The valorisation of coproducts and residues was considered in all integrated biorefineries. MFA was carried out at two levels: the first level concerned the calculation of released sugars and the formation of inhibitors to identify the bottlenecks of the process; the second level concerned the quantification of bioethanol production, and of water, energy and chemicals consumption and of waste generation.

2.2 Economic assessment

The itemised costs involved in the economic assessment of plant design are detailed in Table 1. Capital and operational costs, revenues, net present value (NPV), return of investment (ROI) and payback time were considered in the assessment. The cost of the land was excluded by the capital cost since the study was not geo-referred. For the capital costs, amortisation with 2 % of interest for 5 years in 20 years plant lifetime was considered (Demichelis et al., 2018). The economic profitability was reached when revenues, NPV and ROI were positive and payback time was shorter than plant life. The price of bioethanol with 99.7 % v/v purity was set to 0.91 €/kg, while for the co-products to 0.015 €/kg for vinasse from sugarcane and to 0.017 €/kg for dried distilled grains in soluble (DDGS) from potatoes (PubChem, 2018).

Table 1. Details of the considered plant costs

equipment	unit	cost [€/unit]	Reference
grinder	kg/s	2323.3	(Akerberg and Zacchi, 2000)
reactor	m^3	2514.7	(Dennehy et al., 2017)
stirrer	kw	46465.3	(Akerberg and Zacchi, 2000)
centrifuge	kg/s	116163.2	(Akerberg and Zacchi, 2000)
micro-filter membrane and module	m^2	24637.8	(Akerberg and Zacchi, 2000)
ultrafilter membrane and module	m^2	6164.1	(Akerberg and Zacchi, 2000)
dryer	no.	1152.3	(Akerberg and Zacchi, 2000)
electrodialysis unit	no.	6576.3	(Akerberg and Zacchi, 2000)
electrode	no.	3288.2	(Akerberg and Zacchi, 2000)
heat exchanger	m^2	889.96	(Akerberg and Zacchi, 2000)

2.3. Environmental assessment

The environmental analysis considered the technically feasible and economically profitable configurations for bioethanol fermentative production. The environmental assessment was performed through 1) Life Cycle Analysis (LCA) according to the ISO 14040/44 framework by means of Open

LCA software and 2) energy balance. The geographical origin of the biomass categories (sugar, starch and lignocellulosic) were not considered to avoid inconsistency.

LCA goal and scope were: 1. the evaluation of the potential environmental impacts of bioethanol production from different biomass categories and biorefineries; 2. on the base the outcomes, to define the process outline having the lowest impacts and to underline the pro and cons and bottlenecks. The LCA of the fermentative conversion of bioethanol from sugar, starch and lignocellulosic biomasses was performed considering two functional units (FU), 1 ton of biomass and 1000 L of bioethanol, to prove the consistency of the environmental impact outcomes and to consider two complementary perspectives: biomass valorisation and production of bioethanol. A cradle-to-gate system was defined and the boundaries were: feedstocks transportation from land to plant area, biomass preparation and biomass conversion to bioethanol.

LCI, life cycle inventory: the inventory data were calculated from primary and secondary sources (see Supplementary Materials, Table I). Secondary sources were reviews and scientific papers from Science Direct and Scopus databases. Feedstock transport distance from land to plant area was assumed equal to 50 km and EURO5 diesel trucks were hypothesized to cover the route. The feedstock preparation involved mechanical operations as milling, mixing, drying, chopping etc. The LCA study was performed in EU-28 and based on EU energy mix in 2016.

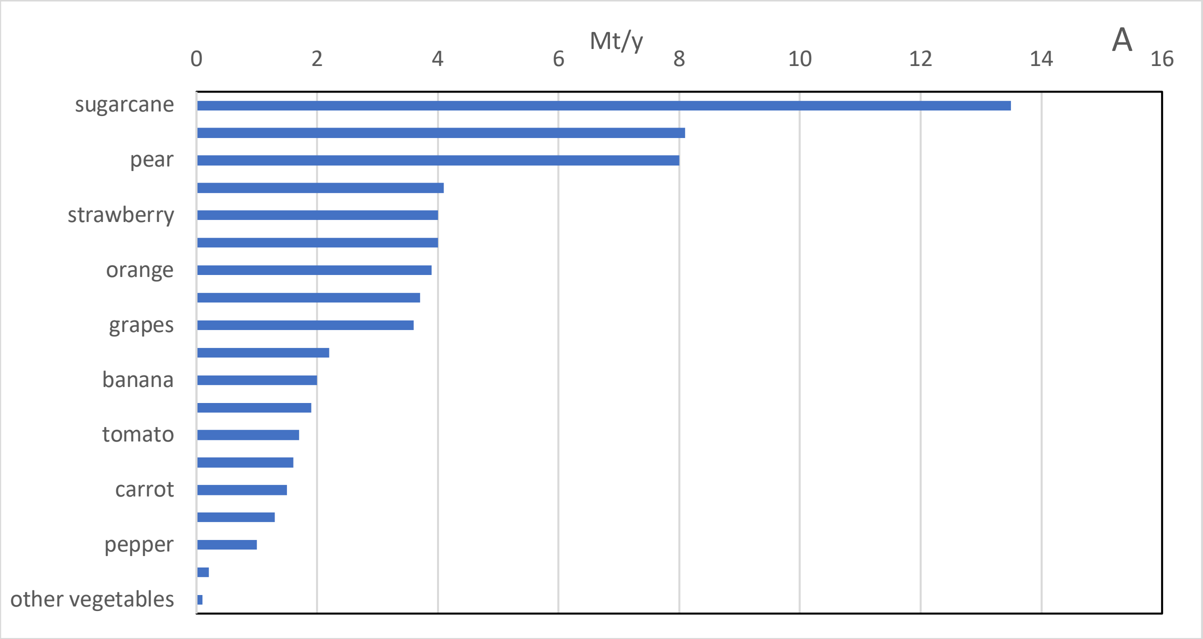
LCIA, life cycle impact assessment: the LCA was performed using Open LCA 1.7 software according to ReCiPe midpoint method. The following categories (and units) were considered (see Supplementary Materials, Table II): Climate change (CC, kg CO_2eq), photochemical oxidation formation (POF, kg NMVOC), particulate matter (PM, kg PM10 eq), terrestrial acidification (TA, kg SO_2 eq), fresh water eutrophication (FEW, kg P eq), marine eutrophication (ME, kg N eq), terrestrial ecotoxicity (TE, kg1,4-DBeq) and fossil depletion (FD, kg oil eq).

The energy evaluation adopted the same FUs employed for LCA (1t of biomass and 1000L of bioethanol) and it was performed for each specific step and for the whole process. The following energy indicators were calculated: net energy value (NEV, e.g. the difference between total energy outputs and inputs), and the net energy ratio (NER, e.g. the ratio between net energy outputs/inputs). The process simulation technique was integrated into LCA and energy evaluations to reduce biased parameters in process data collection (Rathnayake et al., 2018).

3. RESULTS

3.1. Technical assessment

In 2016, bioethanol production in EU-28 from renewable feedstock derived from 71 % starch-based biomass (3.9 Mt from wheat, 4.1 Mt from maize, 0.4 Mt from barley and 0.4 Mt from rye), 24 % sugar-based biomass and 5 % lignocellulosic-based biomass (European Biomass Industry Association, 2018; European Commission, 2018). The inventory of waste biomasses produced in EU-28 according to international databases (Eurostat, 2016a, 2016b, 2016c; FAO, 2018; European Commission, 2012) involved three categories: sugar (Figure 1A), starch (Figure 1B) and lignocellulosic (Figure 1C). The net amounts of each category were calculated subtracting from the specific produced biomass amounts the average losses occurring during harvesting (6%), transport (30%) and processing (20%) (Rezaei and Liu, 2017). The most abundant available biomass category in EU-28 was lignocellulosic (329.41 Mt/y), followed by starch (160 Mt/y) and sugar (58.56 Mt/y).



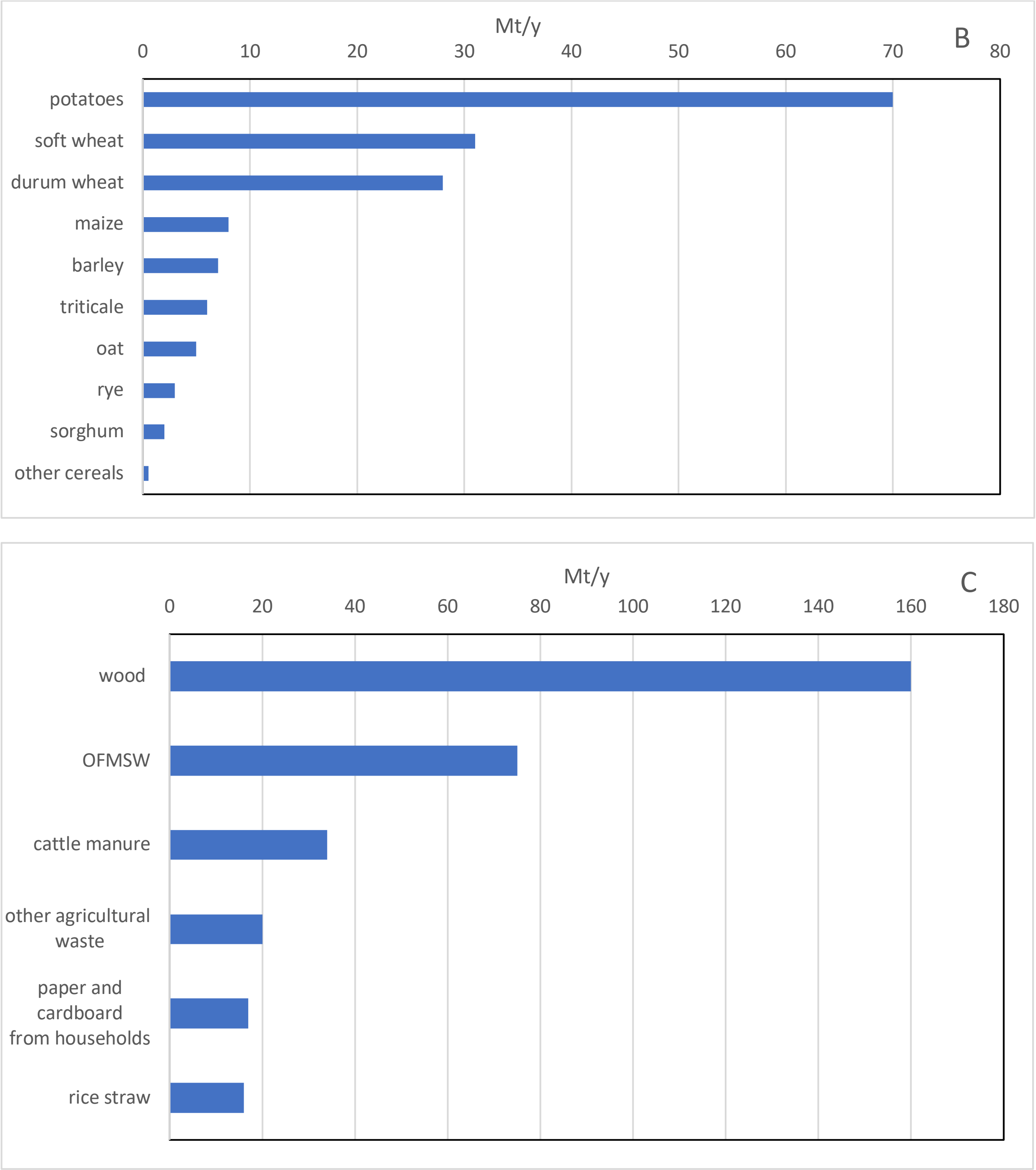


Figure 1. Inventory of waste biomasses (Mt/y) available in EU-28: (A) sugar, (B) starch and (C) lignocellulosic

The qualitative analysis involved only the most abundant biomass of each category referred in the scientific literature to a biorefinery process for the conversion into bioethanol. According to this criterion, the biomasses selected for the TEEA were sugarcane for sugar category; potatoes for starch category; rice straw, cattle manure and OFMSW for lignocellulosic category. Their physico-chemical features are described in Figures 2A (elemental composition) and 2B (nutritional composition) (Banerjee et al., 2017; Chung et al., 2018; Barone et al., 2018; Venus et al., 2018).

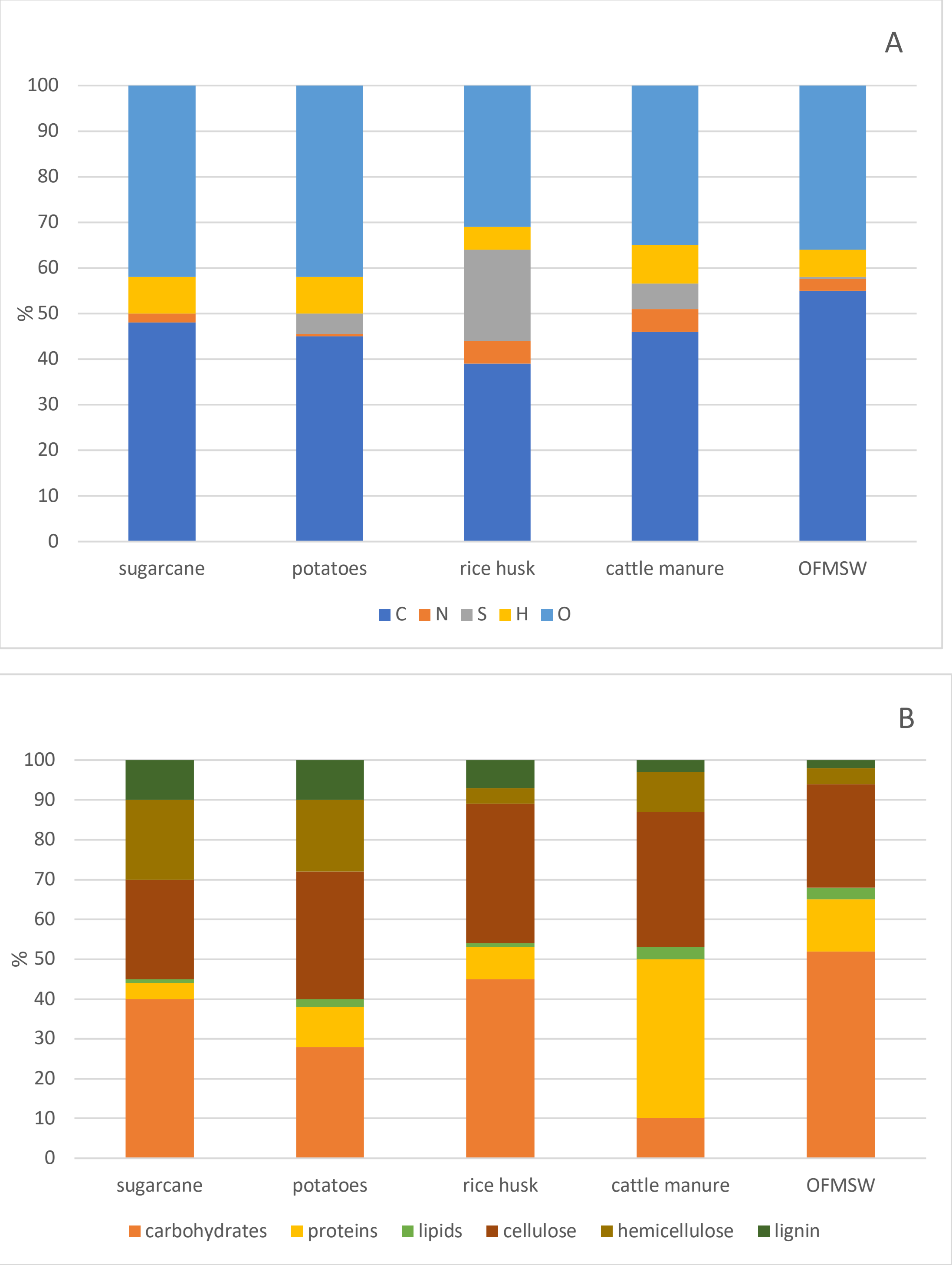
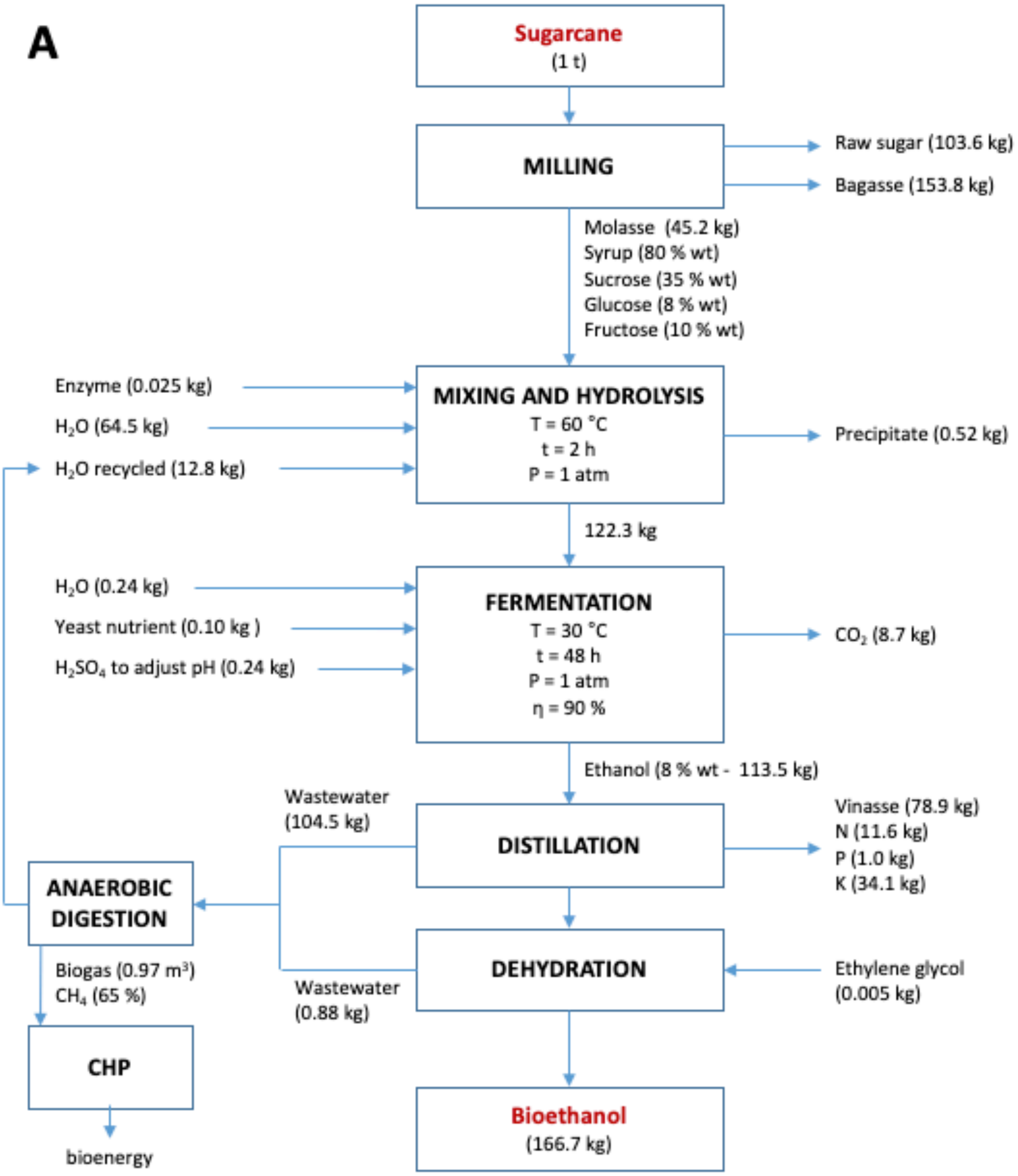
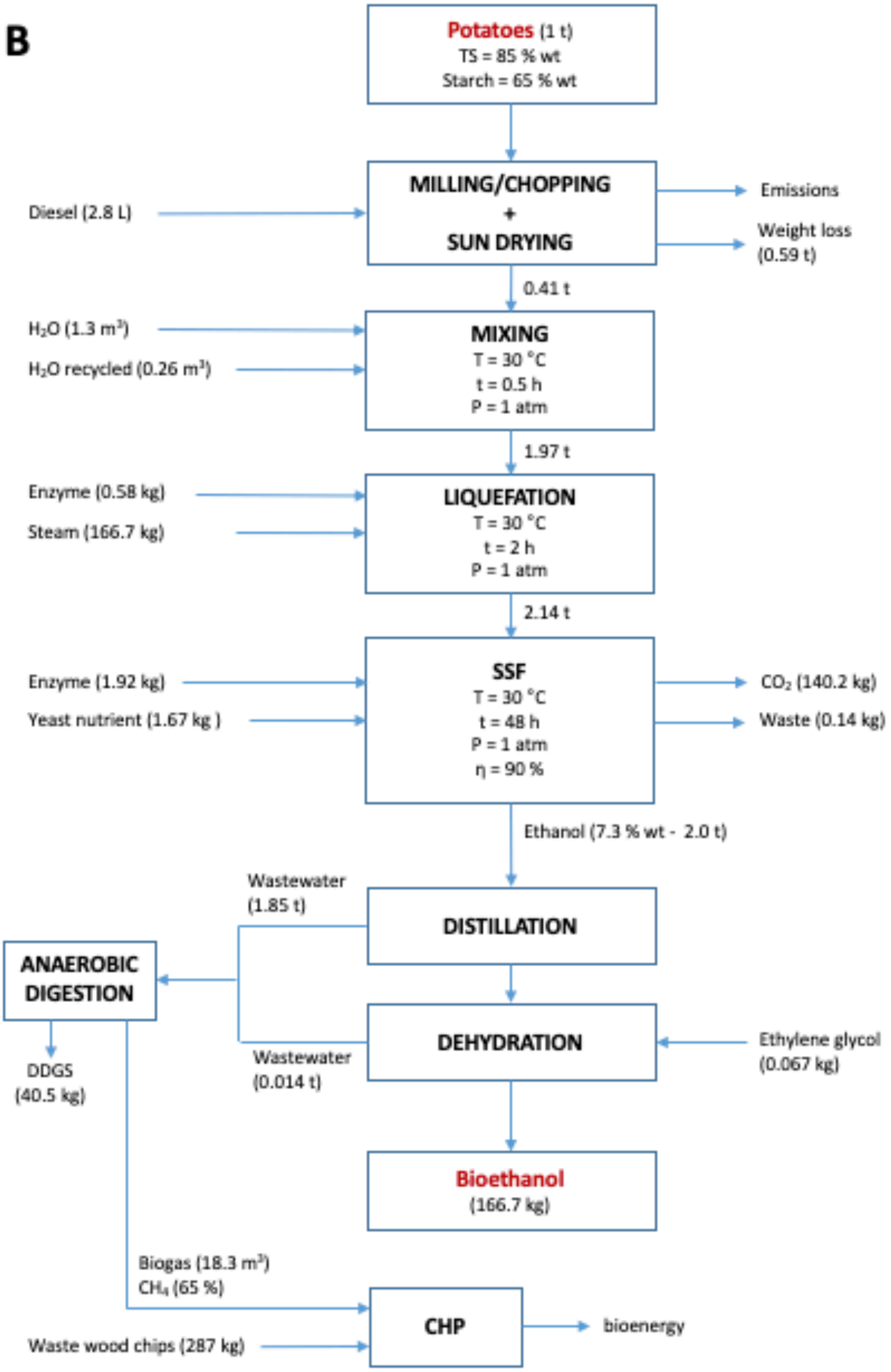
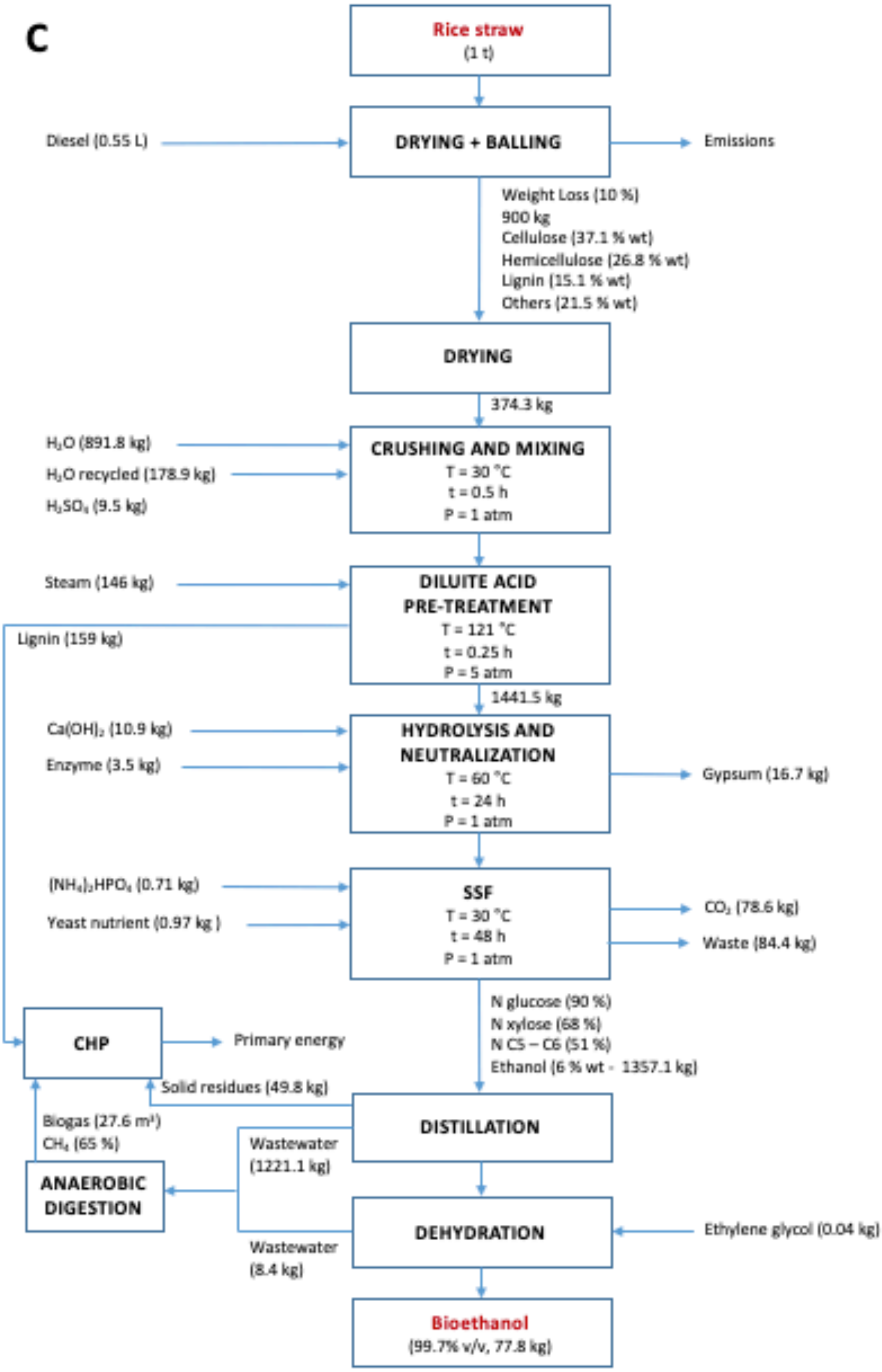


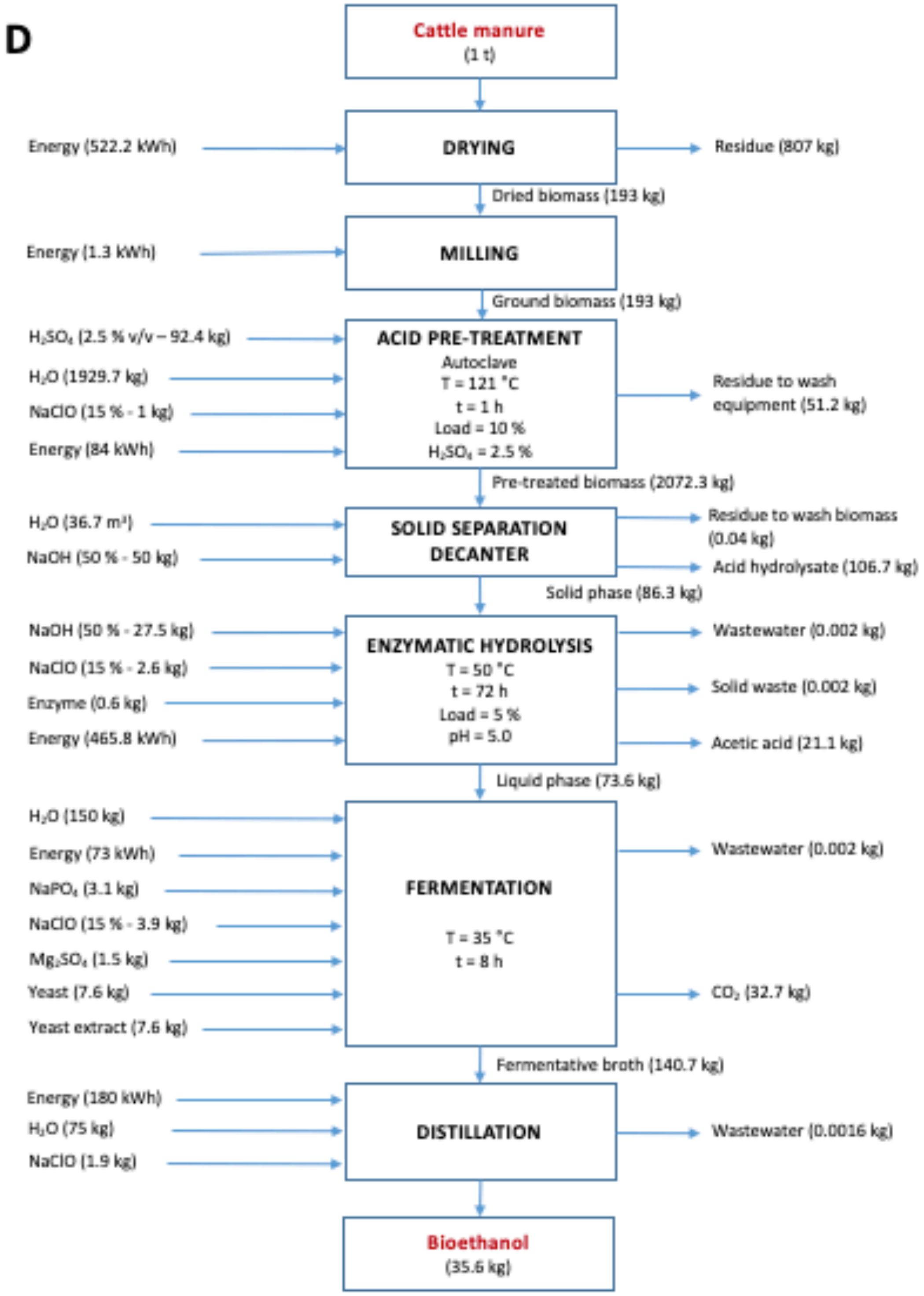
Figure 2. Qualitative description of the selected waste biomasses: (A) elemental and (B) nutritional composition

Almost all the selected biomasses exhibited carbon content exceeding 45-50 %-wt, making them suitable feedstock for biorefinery. The lowest carbon content (38 %-wt) was recorded for rice straw, however the value is appropriate for bioethanol production. The total solid (TS) and volatile solid (VS) contents witnessed the high amount of available organic matter: TS and VS were respectively 77 %-wt and 98 %-wt for sugarcane, 89 % %-wt and 94 % %-wt for potatoes (Banerjee et al., 2017), 88 % %-wt and 98 % %-wt for rice straw (Chung et al., 2018), 19 %-wt and 98 %-wt for cattle manure (Barone et al., 2018) and 20 %-wt and 88 %-wt for OFMSW (Venus et al., 2018). All biomasses displayed significant carbohydrate content (40-70 %-wt), which makes them an appropriate feedstock for the conversion into bioethanol. Only cattle manure had lower carbohydrate content (about 10 %-wt), with high amount of cellulose and hemicellulose. Based on literature data, lignocellulosic biomass exhibited the highest bioethanol yield (450-510 L/t) (Zabed et al., 2017; Zhao et al., 2018), followed by starch-based biomass (365-535 L/t) (Chen and Khanna, 2018; Veljković et al., 2018) and finally by sugar-based biomass (70-107 L/t) (Goldemberg and Guardabassi, 2010). According to the above-cited literature references, bioethanol yields depend on biomass quality (specifically the sugar content and its release during the biological conversion), since bioethanol production is based on glucose conversion by means of yeast or microorganisms. The results accomplished by the qualitative inventory were: sugar-based biomass as sugarcane was mainly composed of sucrose (disaccharide); starch-based biomass after liquefaction and saccharification mainly released glucose (monosaccharide) and maltose (disaccharide); lignocellulosic-based biomasses were composed of 32 -54 %-wt cellulose (Zhao et al., 2018), which released glucose, and 11 – 37 % hemicellulose (Zhao et al., 2018), which released pentose (C5, not readily fermentable sugar) and hexose (C6, readily fermentable). Thus, the detailed knowledge of biomass composition was fundamental in this work for the subsequent design of the integrated biorefinery processes and to achieve the highest yield of bioethanol. After the setup of the quantitative and qualitative inventory of biomasses, the MFA of the bioethanol production was performed, considering 1t of biomass as FU. The target of the MFA was to specifically identify the amounts of the items involved in the integrated biorefinery: sugars deriving from the biomasses, by-products and wastes, chemicals and energy consumed, and bioethanol produced (see Figure 3 A-E).




B
Potatoes (1 t)
TS = 85 % wt
Starch = 65 % wt
MILLING/CHOPPING
+
SUN DRYING
Diesel (2.8 L)
Emissions
Weight loss (0.59 t)
0.41 t
MIXING
T = 30 °C
t = 0.5 h
P = 1 atm
H₂O (1.3 m³)
H₂O recycled (0.26 m³)
1.97 t
LIQUEFATION
T = 30 °C
t = 2 h
P = 1 atm
Enzyme (0.58 kg)
Steam (166.7 kg)
2.14 t
SSF
T = 30 °C
t = 48 h
P = 1 atm
η = 90 %
Enzyme (1.92 kg)
Yeast nutrient (1.67 kg)
CO₂ (140.2 kg)
Waste (0.14 kg)
Ethanol (7.3 % wt - 2.0 t)
DISTILLATION
Wastewater (1.85 t)
DEHYDRATION
Wastewater (0.014 t)
Ethylene glycol (0.067 kg)
ANAEROBIC DIGESTION
DDGS (40.5 kg)
Bioethanol (166.7 kg)
Biogas (18.3 m³)
CH₄ (65 %)
CHP
Waste wood chips (287 kg)
bioenergy


C
Rice straw
(1 t)
Diesel (0.55 L)
DRYING + BALLING
Emissions
Weight Loss (10 %)
900 kg
Cellulose (37.1 % wt)
Hemicellulose (26.8 % wt)
Lignin (15.1 % wt)
Others (21.5 % wt)
DRYING
374.3 kg
H_2O (891.8 kg)
H_2O recycled (178.9 kg)
H_2SO_4 (9.5 kg)
CRUSHING AND MIXING
T = 30 °C
t = 0.5 h
P = 1 atm
Steam (146 kg)
DILUITE ACID
PRE-TREATMENT
T = 121 °C
t = 0.25 h
P = 5 atm
Lignin (159 kg)
1441.5 kg
$Ca(OH)_2$ (10.9 kg)
Enzyme (3.5 kg)
HYDROLYSIS AND
NEUTRALIZATION
T = 60 °C
t = 24 h
P = 1 atm
Gypsum (16.7 kg)
$(NH_4)_2HPO_4$ (0.71 kg)
Yeast nutrient (0.97 kg)
SSF
T = 30 °C
t = 48 h
P = 1 atm
CO_2 (78.6 kg)
Waste (84.4 kg)
N glucose (90 %)
N xylose (68 %)
N C5 – C6 (51 %)
Ethanol (6 % wt - 1357.1 kg)
CHP
Primary energy
Solid residues (49.8 kg)
DISTILLATION
Biogas (27.6 m³)
CH_4 (65 %)
Wastewater
(1221.1 kg)
ANAEROBIC
DIGESTION
DEHYDRATION
Ethylene glycol (0.04 kg)
Wastewater
(8.4 kg)
Bioethanol
(99.7% v/v, 77.8 kg)


D
Cattle manure
(1 t)
Energy (522.2 kWh)
DRYING
Residue (807 kg)
Dried biomass (193 kg)
Energy (1.3 kWh)
MILLING
Ground biomass (193 kg)
H_2SO_4 (2.5 % v/v – 92.4 kg)
H_2O (1929.7 kg)
NaClO (15 % - 1 kg)
Energy (84 kWh)
ACID PRE-TREATMENT
Autoclave
T = 121 °C
t = 1 h
Load = 10 %
H_2SO_4 = 2.5 %
Residue to wash equipment (51.2 kg)
Pre-treated biomass (2072.3 kg)
H_2O (36.7 m^3)
NaOH (50 % - 50 kg)
SOLID SEPARATION DECANTER
Residue to wash biomass (0.04 kg)
Acid hydrolysate (106.7 kg)
Solid phase (86.3 kg)
NaOH (50 % - 27.5 kg)
NaClO (15 % - 2.6 kg)
Enzyme (0.6 kg)
Energy (465.8 kWh)
ENZYMATIC HYDROLYSIS
T = 50 °C
t = 72 h
Load = 5 %
pH = 5.0
Wastewater (0.002 kg)
Solid waste (0.002 kg)
Acetic acid (21.1 kg)
Liquid phase (73.6 kg)
H_2O (150 kg)
Energy (73 kWh)
$NaPO_4$ (3.1 kg)
NaClO (15 % - 3.9 kg)
Mg_2SO_4 (1.5 kg)
Yeast (7.6 kg)
Yeast extract (7.6 kg)
FERMENTATION
T = 35 °C
t = 8 h
Wastewater (0.002 kg)
CO_2 (32.7 kg)
Fermentative broth (140.7 kg)
Energy (180 kWh)
H_2O (75 kg)
NaClO (1.9 kg)
DISTILLATION
Wastewater (0.0016 kg)
Bioethanol
(35.6 kg)

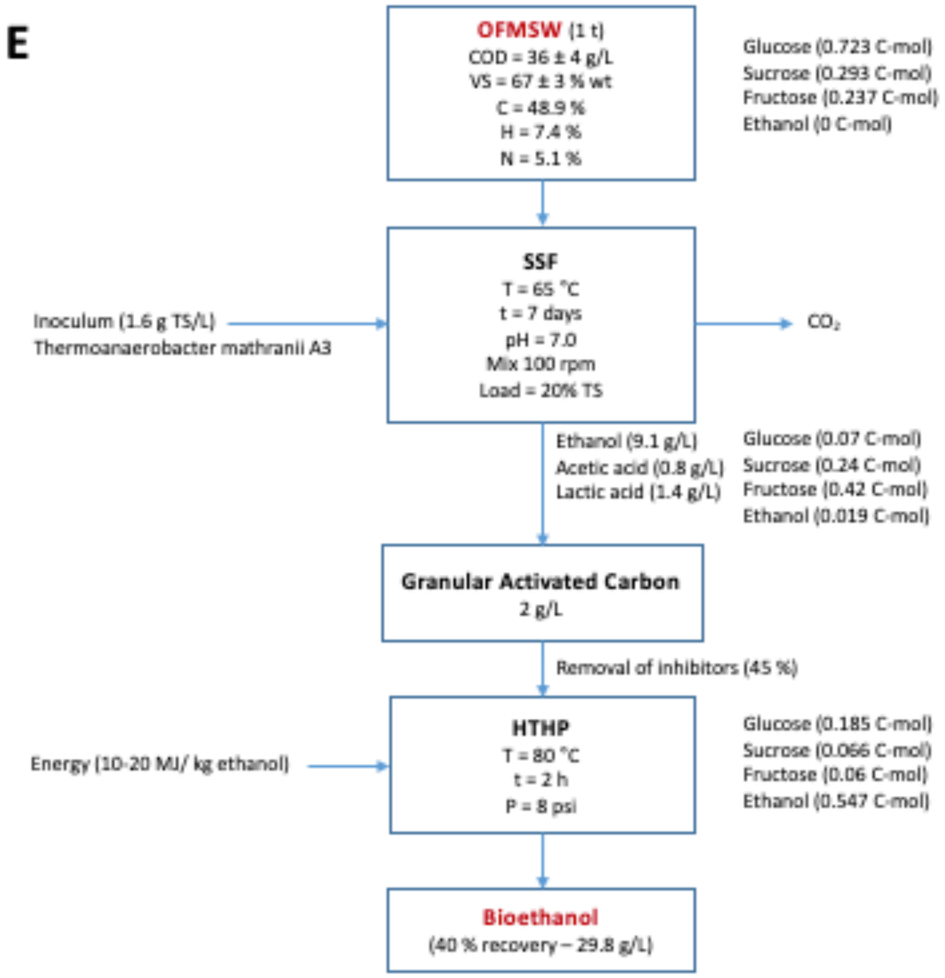


Figure 3. MFA of bioethanol production from (A) sugarcane, (B) potatoes, (C) rice straw, (D) cattle manure and (E) OFMSW

The integrated biorefineries were defined according to the physico-chemical composition the five selected biomasses and to the conversion process outlines available in the scientific literature and referred to different technology readiness level (TRL). Sugarcane (sugar category, Figure 3A), potatoes (starch category, Figure 3B) and rice straw (lignocellulosic category, Figure 3C) were considered within integrated biorefinery system with TRL 7 (Rathnayake et al., 2018), while cattle manure (lignocellulosic category, Figure 3D) (de Azevedo et al., 2017) and OFMSW (lignocellulosic category, Figure 3E) (Barampouti et al., 2019) were involved in simple biorefinery systems, respectively with TRLs 4 and 2. Anaerobic digestion (AD) and a combined heat and power (CHP) unit were included in the flow charts to treat wastewater and recover energy. The outlines of the biorefinery processes considered in the research are described in the following, together with the results of the MFA.

Sugarcane conversion process consisted of 5 steps (Figure 3A) (Rathnayake et al., 2018): milling to obtain molasse, hydrolysis, fermentation, distillation and dehydration. From 1 ton of sugarcane, 45.20 kg of molasses was obtained with 53 %-wt of C6 sugars, ready convertible. From milling and distillation respectively 153.84 kg of bagasse and 78.93 kg of vinasse were produced as coproducts. The chemicals consumption represented 37 %-wt of the FU. The yield was 166.7 kg of bioethanol per kg of sugarcane.

Potatoes conversion process consisted of 5 steps (Figure 3B) (Morales et al., 2015): milling/chopping, drying, enzymatic liquefaction, fermentation, distillation and dehydration. From AD process, DDGS for animal feed was produced as coproduct. After drying, the available waste biomass obtained from 1 ton of potatoes was 41 %-wt. The chemicals consumption represented 47 %-wt of the FU, mostly due to enzymatic liquefaction and fermentation. The yield was 166.7 kg bioethanol per kg of potatoes. The boundary conditions of our calculations gave back the same results for sugarcane and potatoes; even if different by nature (the first is sugar-based, the second starch-based biomass), both feedstocks produce bioethanol from the fermentation of hydrolyzed sugars, as before mentioned.

Rice straw process consisted of 8 steps (Figure 3C) (Morales et al., 2015; Zabed et al., 2017; Zhao et al., 2018): drying, crushing and mixing, enzymatic hydrolysis, fermentation, distillation and dehydration. 37 %-wt of rice straw was made of cellulose, and it required pre-treatment and hydrolysis to release sugar. The chemicals consumption represented 60 %-wt of FU due to pre-treatment and enzymatic liquefaction and fermentation. The yield was 77.8 kg of bioethanol per kg of rice straw.

Cattle manure conversion process consisted of 7 steps (Figure 3D) (Zabed et al., 2017; Zhao et al., 2018): drying, milling, acid pre-treatment, solid separation, enzymatic hydrolysis, fermentation, distillation. The chemical consumption represented 60 – 63 %-wt of FU due to pre-treatment, enzymatic liquefaction and fermentation. The yield was 35.6 kg of bioethanol per kg of cattle manure. OFMSW conversion process consisted of three steps (Figure 3E) (Demichelis et al., 2017; Dhiman et al., 2017; Zhao et al., 2018): simultaneous saccharification and fermentation (SSF), adsorption on granular activated carbon, high temperature and high-pressure (HTHP) downstream processes. During SSF, the production of lactic acid and acetic acid (3.2 g/L and 0.32 g/L respectively) justified the pH drop of the fermentative broth and the relevant employment of chemicals (50%-wt of FU). The yield was 29.8 kg of bioethanol per kg of OFMSW.

According to scientific literature, the highest yield of bioethanol was achieved by sugar- and starch-based biomass (sugarcane and potatoes), followed by lignocellulosic (rice straw, cattle manure and OFMSW). At the same time lignocellulosic feedstocks generated the highest amount of waste and coproducts consuming energy and chemicals throughout the conversion process (Morales et al., 2015).

3.2 Economic evaluation

The economic analysis involved all the five selected biomasses, since for all of them a technically feasible solution was demonstrated. The highest capital and operational costs were reached by cattle manure (Figure 4). The capital cost was due to the requirement of several equipment for pre-treatment and conversion of biomass (Figure 3D), while operational costs were due to waste generation (around 90 %-wt of feedstock) and chemicals employed for the pre-treatments. In general, the highest investment costs were reached by lignocellulosic biomasses because of the higher complexity of process outline due to the need of pre-treatment and downstream steps (Figure 3C-E) (Morales et al., 2015; Sanchez et al., 2016).

About economic profitability (Table 2), the best performance was achieved by sugarcane, followed by potatoes, rice straw, OFMSW and cattle manure. The highest NPV was achieved by sugarcane (because this sugar-based biomass was easily convertible into bioethanol, with the highest yield) (Figure 3A) and thus required lower investment costs, followed by OFMSW. For the minimum necessary size plant to achieve profitability, all biomasses exhibited a payback time close or equal to plant life. Cattle manure reached the lowest values of ROI and NPV, according to literature (Cherubini et al., 2015).

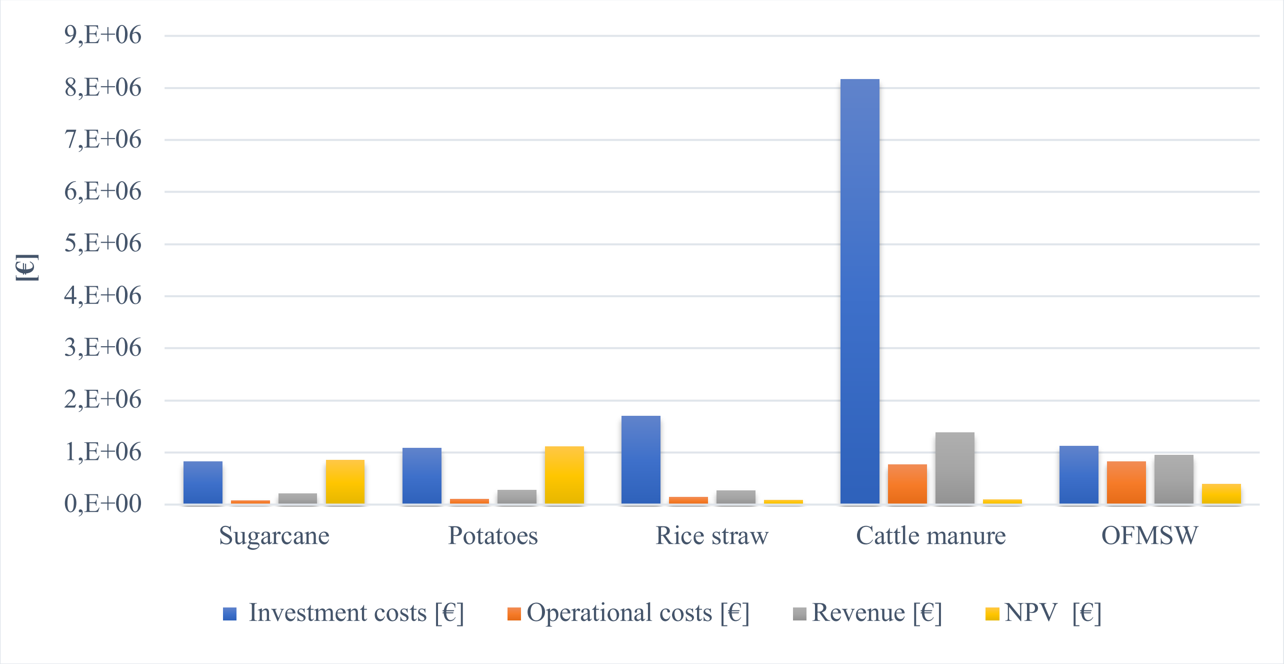


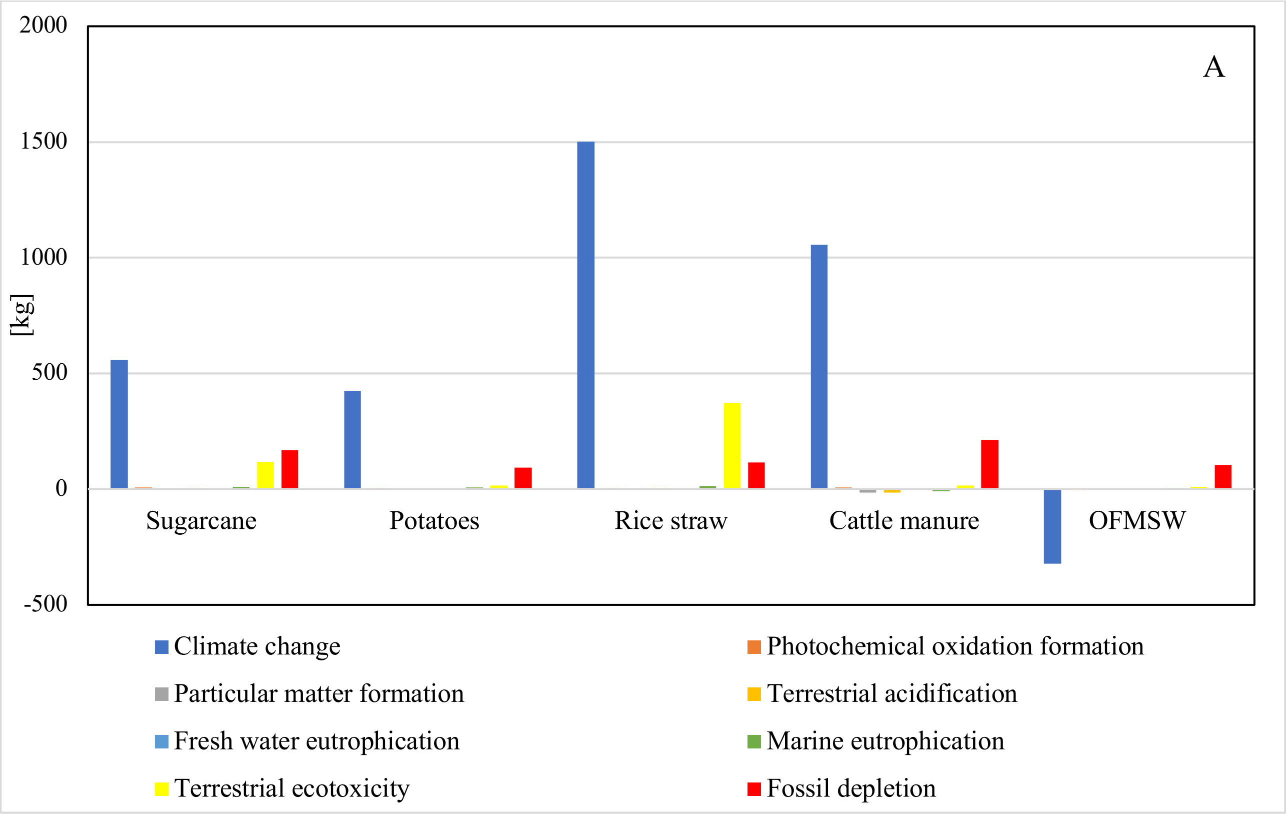
Figure 4. Results of the economic analysis (evaluation of capital and operational costs, revenues and NPV after 5 years) for bioethanol production from sugarcane, potatoes, rice straw, cattle manure and OFMSW

Table 2. Results of the economic assessment of bioethanol production from sugar cane, potatoes, rice, cattle manure and OFMSW based on NPV, ROI and payback time

	Sugarcane	Potatoes	Rice straw	Cattle manure	OFMSW
substrate available [Mt/y]	13.5	70	16	34	75
substrate used [%]	0.011	0.003	0.010	0.027	0.0056
substrate used [t/y]	1450	1900	1700	9000	4250
ROI [%]	15.7	15.6	7.8	7.5	10.1
NPV [M€]	0.85	0.11	0.09	0.11	0.39
Payback time [y]	20.0	20.0	19.0	20.0	18.0

3.3 Environmental evaluation

The results of LCA performed considering as FU 1000 L ethanol (see Figure 5 A and B) and 1 ton of biomass (see Figure 5 C and D) confirmed that the highest impact category was climate change (particularly for rice straw), followed by terrestrial ecotoxicity and fossil depletion. The use of chemicals for pre-treatments, hydrolyses and fermentations contributed to the impact categories climate change, toxicity, fossil depletion and metal deposition and freshwater eutrophication, in agreement with (Benjamin et al., 2014). However, OFMSW conversion to bioethanol positively contributed to the reduction of climate change impact and terrestrial ecotoxicity, in agreement with literature (Nayak and Bhushan, 2019). The only lack of consistency of the LCA results obtained adopting two different FUs was achieved by OFMSW, with the impact category of terrestrial acidification. Among the five considered biomasses, cattle manure and OFMSW achieved the highest saving of environmental impacts followed by potatoes, sugarcane and rice straw. The positive environmental outcomes of cattle manure conversion into bioethanol was mostly due to the reduction of pathogenic emissions causing human health problems and of the environmental impact for nitrogen and phosphorous contamination in soil, as in literature (Junior et al., 2015). In fact, 82 % of total environmental impacts are due to ammonia emission when cattle manure is stored in open tanks, while biogas production from cattle manure released NOx emissions thus reducing SO_2 emissions (Aditiya et al., 2016). Consequently, the LCA performed in this study on cattle manure conversion into bioethanol achieved: for terrestrial acidification -147 kg SO_2 eq (FU 1000 L of ethanol) and -0.21 kg SO_2 eq (FU 1 tonof biomass) and particular matter formation -14.28 kg PM10 eq (FU 1000 of L ethanol) and -0.02 kg PM10 eq (FU 1 ton of biomass).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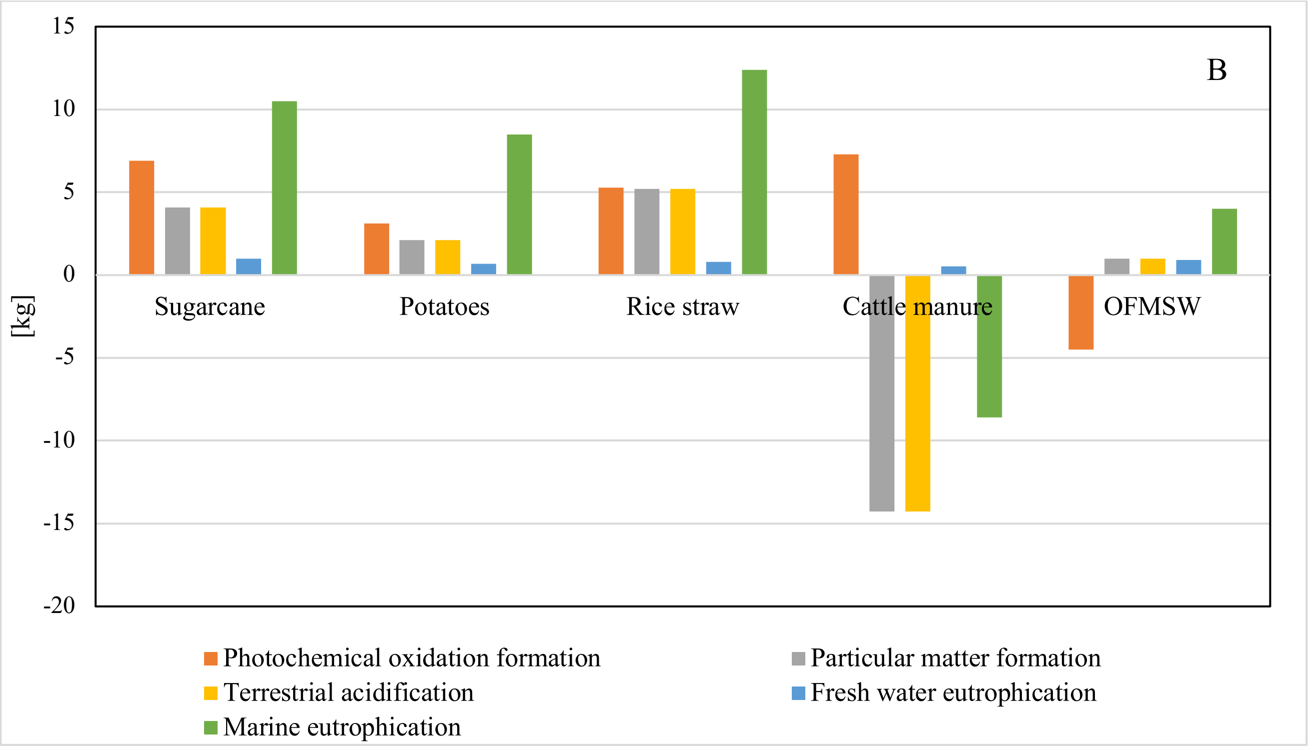
Particular matter formation
Terrestrial acidification
Fresh water eutrophication
Marine eutrophication
Terrestrial ecotoxicity
Fossil depletion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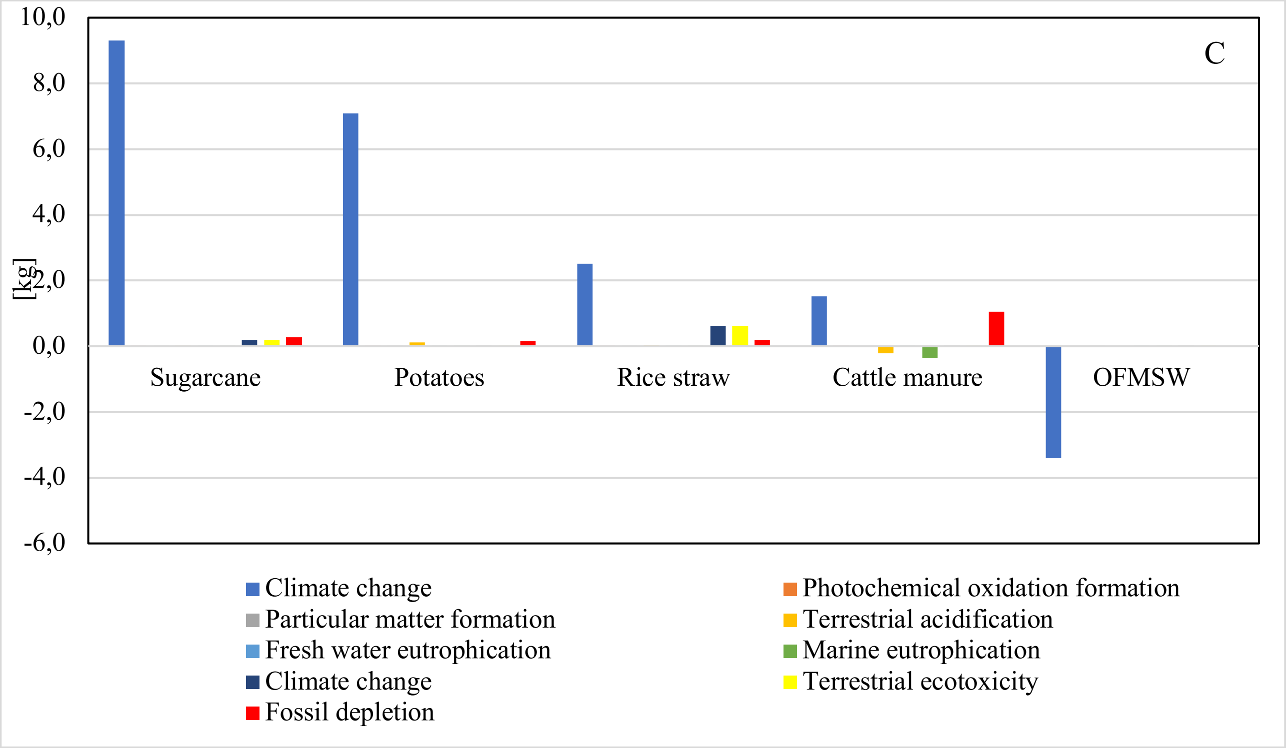

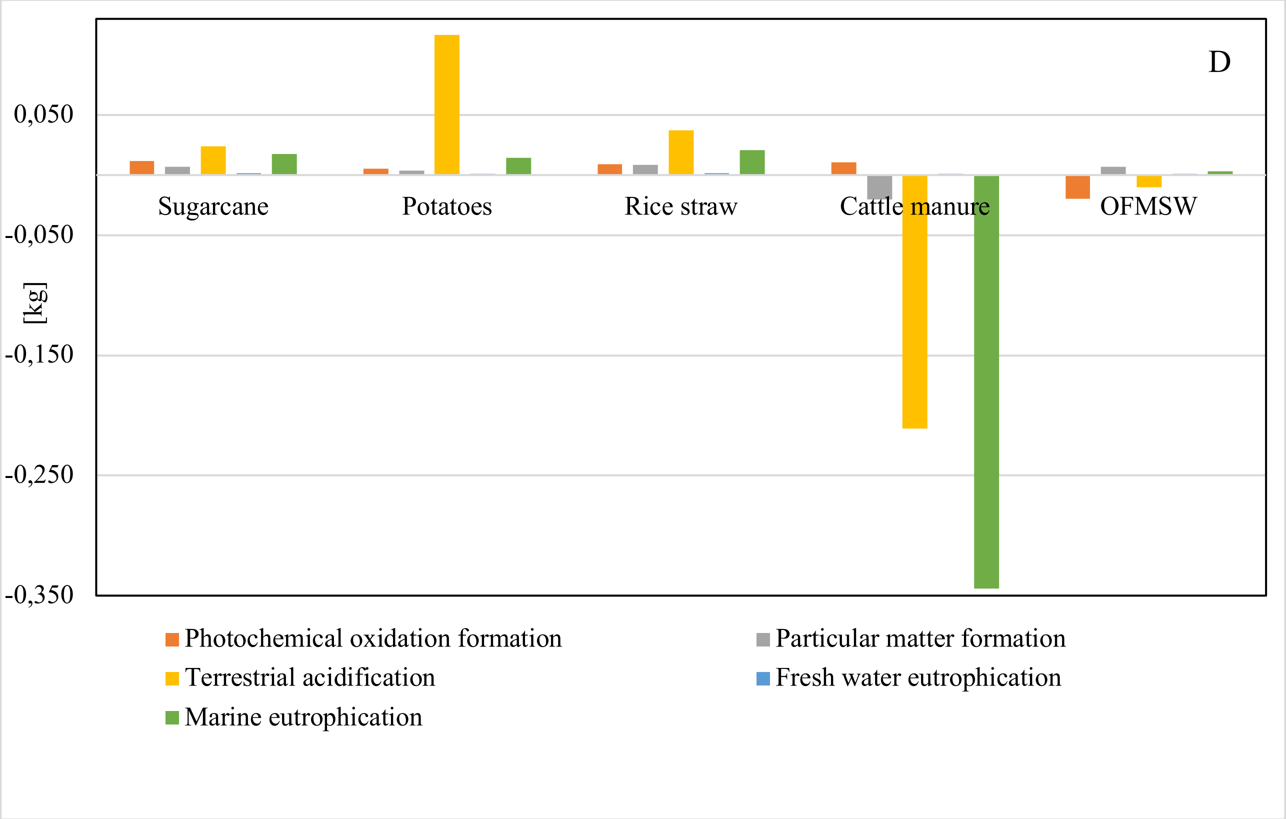

Figure 5. Results of LCA (FU 1000 L of ethanol): A) impact categories CC, TE and FD; B) other impact categories; LCA results (FU 1 ton of biomass): C) impact categories CC, TE and FD; D) other impact categories

The energy assessment (Figure 6) detected that the highest specific energy consumption item for all the considered biorefineries were generally the downstream processes (30-50%), followed by fermentation (10-20%) in agreement with (Demichelis and Fiore, *in preparation*; Rathnayake et al., 2018). Pre-treatments exhibited the widest range of values, from 1.88 % (potatoes) to 81.35 % (cattle manure), depending on the feedstock and specifically about cattle manure related to the high energy request of drying phase, due to its high moisture content (81 %, see section 3.1).

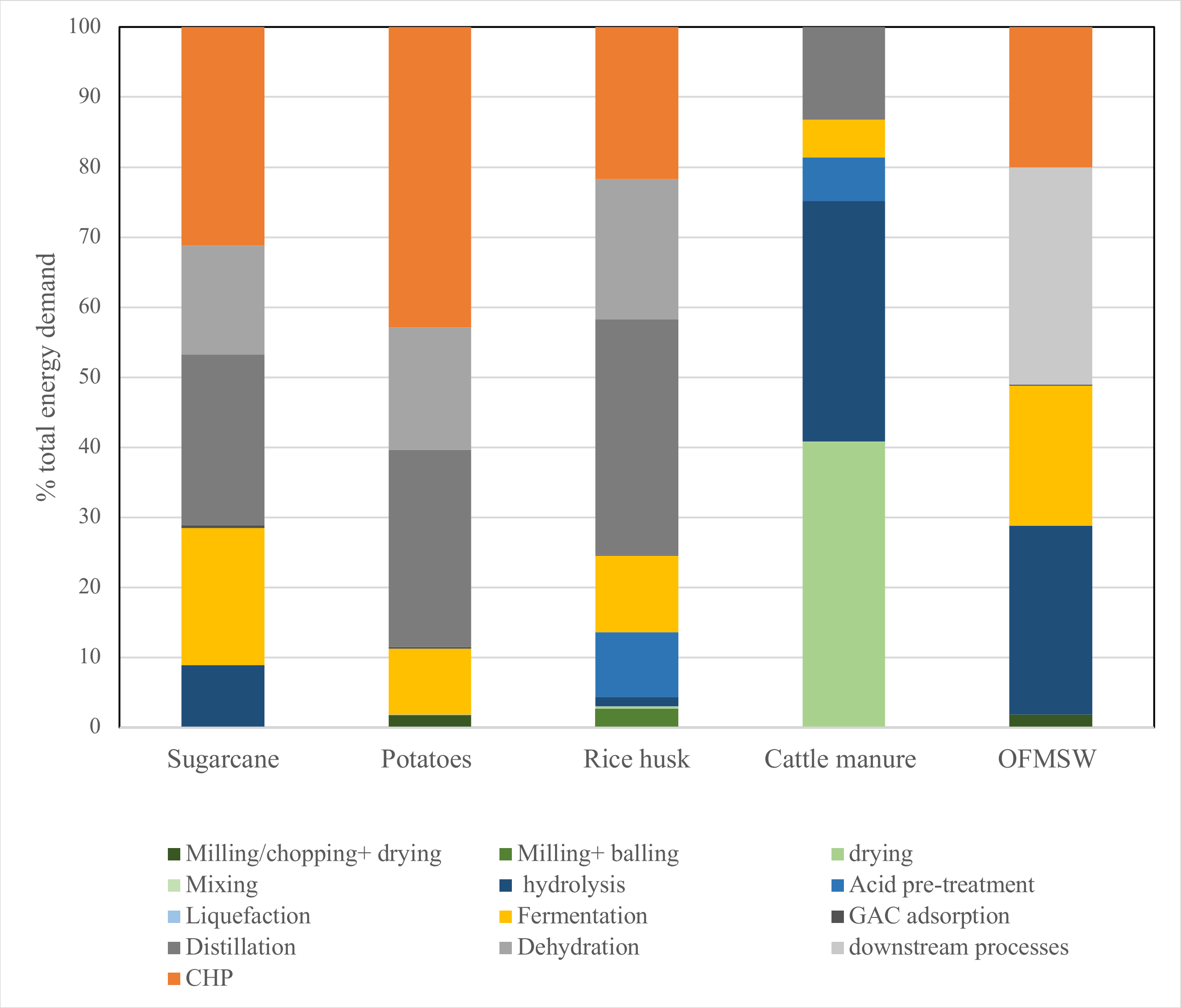

Figure 6. Details of the total energy requirements of the considered biorefineries

Table 3. Parameters and results of energy analysis (FU 1000 L of bioethanol)

	Sugarcane	Potatoes	Rice straw	Cattle manure	OFMSW
Energy consumption pre-treatments [kWh]	2577.2	210.3	70.6	1428.7	
Energy consumption chemicals [kWh]	22.5	71.1	337.8	69.8	
Energy production steam [kWh]	6601.6	3313.9	5837.7	0.0	
Energy production electricity [kWh]	1649.0	918.4	1734.6	0.0	
Energy consumption steam [kWh]	1809.6	2689.1	4852.3	1690.5	
Energy consumption electricity [kWh]	788.1	918.4	1584.3	0.0	
Surplus energy steam [kWh]	2214.9	624.8	985.4	0.0	
Surplus energy electricity [kWh]	794.8	0.0	150.3	0.0	
Total net energy inputs [kWh]	8128.4	4921.5	8277.6	3188.9	6575.5
Total net energy outputs [kWh]	8899.0	6514.1	7025.0	0.0	
Total net bio-energy outputs [kWh]	8899.0	6514.1	7025.0	0.0	
NEV [kWh]	770.6	1592.6	- 1252.6	- 3188.9	0.0
NER [-]	0.3	0.4	0.2	0.0	-

Table 4. Parameters and results of energy analysis (FU 1 ton of biomass)

	Sugarcane	Potatoes	Rice straw	Cattle manure	OFMSW
Energy consumption pre-treatments [kWh]	544.4	44.4	7.0	607.5	
Energy consumption chemicals [kWh]	4.8	15.0	33.3	29.7	
Energy production steam [kWh]	1394.5	700.0	575.4		
Energy production electricity [kWh]	348.3	194.0	171.0		
Energy consumption steam [kWh]	382.3	568.1	478.3	718.8	
Energy consumption electricity [kWh]	166.5	194.0	156.2		
Surplus energy steam [kWh]	467.9	132.0	97.1		
Surplus energy electricity [kWh]	167.9	0.0	14.8		
Total net energy inputs [kWh]	1717.1	1039.6	815.9	1356.0	3142.8
Total net energy outputs [kWh]	1879.9	1376.1	692.4		
Total net bio-energy outputs [kWh]	1879.9	1376.1	692.4		
NEV [kWh]	162.8	336.4	- 123.5	- 1356.0	- 3142.8
NER [-]	1.1	1.3	0.8	0.0	-

Taking into account the energy analysis of the whole integrated biorefinery systems, the results of the assessment considering the two complementary FUs (Tables 3 and 4) were consistent. The total energy consumption values related to the considered biorefineries calculated in the present study were in agreement (even if slightly lower) with literature. The literature data concerning energy consumption were: 10000 - 20000 MJ for potatoes, 15000 - 27000 MJ for sugarcane and 10000 -

30000 for rice straw (Le et al., 2013). The highest energy input calculated in this work was achieved by rice straw (29797 MJ equal to 8277.6 kWh), followed by sugarcane (29260 MJ equal to 8128.4 kWh) and potatoes (17716 MJ equal to 4921.5 kWh). The energy consumption was therefore equal to 64 %, 73 % and 79 % of total energy inputs respectively for sugarcane, potatoes and rice straw. These results are consistent with the increase of complexity of the biomass structure passing from sugar- to starch- to lignocellulosic-based feedstock (Quintero and Cardona, 2011). For cattle manure the highest energy item was drying (41 % of total energy consumption, since cattle manure exhibited 18 % TS), followed by hydrolysis (34.5 % of total energy consumption). Considering energy indicators, sugarcane and potatoes ensured net energy gains, while rice straw had negative ones. Energy indicator could not be calculated for cattle manure and OFMSW, since necessary data were not available.

The results of LCA and energy analyses achieved by this study with both FUs were coherent. In details, the energy evaluation observed positive NEV and NER for integrated biorefineries, as for potatoes and sugarcane, while rice straw exhibited an energy loss, since pre-treatments and distillation require high energy rate. According to literature (Saga et al., 2010) bioethanol conversion and purification was the highest item costs, accounting for about 60 - 80 % of total energy.

4. Conclusions

The application of the proposed TEEA methodology for the assessment of bioethanol production from waste biomass allowed to obtain consistent results through the whole procedure of technical, economic and environmental assessment (even considering two different FUs). The considered biomasses presented appropriate qualitative features (at least 40 % carbon and 40 - 70 % carbohydrates) to be considered suitable feedstock for bioethanol production. The bioethanol yields (kg bioethanol /kg feedstock) were: 166.7 for sugarcane and potatoes, 77.8 for rice straw, 35.6 for cattle manure and 29.8 for OFMSW. The economic profitability was reached by all biomasses and the ROI (%) and NPV (M€) were: 15.7 and 0.85 for sugarcane; 15.6 and 0.11 for potatoes; 7.8 and

0.09 for rice straw; 7.5 and 0.11 for cattle manure; 10.1 and 0.39 for OFMSW. The minimum plant size based on economic profitability defined a consumption of the available biomass equal to 1450, 1900, 1700, 9000 and 4250 t/y respectively for sugarcane, potatoes, rice straw, cattle manure and OFMSW. From the environmental perspective, cattle manure allowed the highest reduction of the impacts related to climate change and acidification, while sugarcane achieved the lowest energy input consumption (64 %).

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SUPPLEMENTARY MATERIALS

Table I. Life cycle inventory data of the considered biorefineries (transport was assumed for 50 km through EURO5 trucks for all biomasses, considering 5 tons truckloads. Italian energy mix referred to 2018 was considered)

Reference: (Rathanayake et al., 2018)						
INPUT	unit	amount	**OUTPUT**	unit	amount	destination
SUGARCANE	kg	1000	**bioethanol**	**kg**	**166.7**	
energy for pre-treatments and hydrolyisis	kWh	544.4	bagasse	kg	153.8	recycling
water	L	64.5	precipitate	kg	0.52	recycling
water recycled from anaerobic digestion	L	12.8	wastewater	L	105.4	anaerobic digestion
yeast	kg	0.1	biogas	m3	0.97	bioenergy production
sulfuric acid	kg	0.24	bioenergy produced	kWh	1879.9	
energy for fermentation and downstream processes	kWh	1172.7	vinasse	kg	79.8	recycling
			ethylene glycol	kg	0.005	recycling
Reference: (Morales et al., 2015)						
INPUT	unit	amount	**OUTPUT**	unit	amount	destination
POTATOES	kg	1000	**bioethanol**	**kg**	**166.7**	
energy for pre-treatments and hydrolysis	kWh	44.4	fermentation residues	kg	0.14	recycling
water	L	1300	DDGS	kg	40.5	recycling
water recycled from anaerobic digestion	L	260	wastewater	L	1864	anaerobic digestion
yeast	kg	1.67	biogas	m3	18.3	bioenergy production
enzyme	kg	1.67	bioenergy produced	kWh	1376.1	
energy for fermentation and downstream processes	kWh	995.2	ethylene glycol	kg	0.067	recycling
References: (Morales et al., 2015; Zabed et al., 2017; Zhao et al., 2018)						
INPUT	unit	amount	**OUTPUT**	unit	amount	destination
RICE STRAW	kg	1000	**bioethanol**	**kg**	**77.8**	
energy for pre-treatments and hydrolyisis	kWh	7	fermentation residues	kg	84.4	recycling
water	L	891	wastewater	L	1230	anaerobic digestion
water recycled from anaerobic digestion	L	179	biogas	m3	27.6	bioenergy production
yeast	kg	0.97	bioenergy produced	kWh	1376.1	
enzyme	kg	3.5	ethylene glycol	kg	692.4	recycling
sulfuric acid + other chemicals	kg	21.11				

energy for fermentation and downstream processes	kWh	809				
References: (Zabed et al., 2017; Zhao et al., 2018)						
INPUT	unit	amount	**OUTPUT**	unit	amount	destination
CATTLE MANURE	kg	1000	**bioethanol**	**kg**	**35.6**	
energy for pre-treatments and hydrolyisis	kWh	748.5	fermentation residues	kg	20	recycling
water	L	5825	wastewater	L	2000	treatment
yeast	kg	15.2				
enzyme	kg	0.6				
sulfuric acid + other chemicals	kg	183.9				
energy for fermentation and downstream processes	kWh	749				
References: (Demichelis et al., 2017; Dhiman et al., 2017; Zhao et al., 2018)						
INPUT	unit	amount	**OUTPUT**	unit	amount	destination
OFMSW	kg	1000	**bioethanol**	**kg**	**29.8**	
total energy demand	kWh	3142.8	fermentation residues	kg	822	recycling
chemicals	kg	500	lactic acid	L	148.2	recycling

Table II. Life cycle impact (LCI) categories, units and results of LCI assessment

FU 1000 L of bioethanol						
Impact category	Sugarcane	Potatoes	Rice straw	Cattle manure	OFMSW	unit
Climate change	558,1	425,1	1501,9	1057	-322,56	kg CO2 eq
Photochemical oxidation formation	6,9	3,1	5,3	7,28	-4,5	kg NMVOC
Particular matter formation	4,1	2,1	5,2	-14,28	1	kg PM10 eq
Terrestrial acidification	14,3	7	22,2	-147,7	-4,89	kg SO2
Fresh water eutrophication	1	0,7	0,8	0,5404	0,9	kg N eq
Marine eutrophication	10,5	8,5	12,4	-8,6	4	kg N eq
Terrestrial ecotoxicity	117,3	15	371,8	16,8	10	kg1,4-DB eq
Fossil depletion	168,1	93,7	116	212	103	kg oil eq

FU 1 t of biomass						
Impact category	Sugarcane	Potatoes	Rice straw	Cattle manure	OFMSW	unit
Climate change	9,302	7,085	2,503	1,510	-3,400	kg CO2 eq
Photochemical oxidation formation	0,012	0,005	0,009	0,010	-0,020	kg NMVOC
Particular matter formation	0,007	0,004	0,009	-0,020	0,007	kg PM10 eq
Terrestrial acidification	0,024	0,117	0,037	-0,211	-0,010	kg SO2
Fresh water eutrophication	0,002	0,001	0,001	0,001	0,001	kg N eq
Marine eutrophication	0,018	0,014	0,021	-0,344	0,003	kg N eq
Terrestrial ecotoxicity	0,196	0,025	0,620	0,024	0,010	kg1,4-DB eq
Fossil depletion	0,280	0,156	0,193	1,060	0,240	kg oil eq