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Country report

Environmental comparison of banana waste valorisation strategies under a biorefinery approach

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A B S T R A C T

Banana wastes can be valorised in bioethanol due to its high content in cellulose (more than 30% of total on a dry basis) and hemicelluloses (25% of total). Large amount of these wastes is generated during the banana cultivation and harvesting stage. This study proposes the use of, beside conventional acid sulphuric, different organic acids (tartaric, oxalic and citric) during acid pretreatment step, to suppress the unwanted compounds formation and improve bioethanol production. Instead, bioethanol production generates a solid waste flow that is managed in an anaerobic digestion plant, obtaining biogas, to be converted into energy, and digestate, considered as a potential biofertiliser. Life cycle assessment methodology is used to analyse the environmental profiles of four valorisation scenarios to produce bioethanol from banana peel waste. According to the results, reported per kilogram of bioethanol, the citric acid-based scenario has the worst environmental profile due to the background processes involved in the acid production (around 55% for most impact categories). Conversely, the oxalic acid-based scenario has the best environmental profile, with a decrease of around 20% and 35%, depending on the impact category, compared to the citric acid scenario. The energy requirements production (mostly thermal energy) is the main hotspot in numerous subsystems regardless of the scenario (ranging from 30% to 50% depending on the impact category). Therefore, the use of renewable energy sources to satisfy energy requirements combined with an energy optimisation of the valorisation strategies through the reuse of some internal steams, is proposed as improvement activities.

1. Introduction

According to the FAO statistics, around 33% of food produced globally is lost or wasted along the supply chain (FAO, 2013). Sustainable Development Goal 12 (SDG 12), entitled “Responsible Consumption and Production”, purposes, among other, to implement recycling and reuse strategies in production and supply chains to reduce food losses (United Nations, 2015). Considering the constraint of landfill disposal of food waste according to Council Directive 2008/98/EC (European Union, 2008), other techniques such as incineration or composting are arising for food waste management (Lin et al., 2013). Nevertheless, the content of various components (e.g., lipids, pectins or phytochemicals) present in these wastes emerges as an alternative to recover valuable resources.

Therefore, along with the interest in reducing the amount of food waste, the biorefinery concept aims at converting biomass into high value-added products that can even be commercially exploitable (Cherubini, 2010), such as biofuels (Algapani et al., 2019), biofertilisers (Vico et al., 2018), proteins or lipids (Capellini et al., 2017; Hojilla-Evangelista et al., 2017; Wang et al., 2007) or bioactive compounds as a source of functional ingredients. Depending on the desired final product, as well as the quality, quantity and composition of biomass, there are several parameters that determine the choice of the production process in a biorefinery (Tursi, 2019).

Vegetable-based biomass, characterised by its cellulose (40–50%), hemicellulose (25–35%) and lignin (15–20%) content, although it varies according to its origin, is a source of fermentable sugars in bioethanol production (Danmaliki et al., 2016; Tursi, 2019). Cellulose, a structural

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Abbreviations: AD, Anaerobic Digestion; CHP, Cogeneration Heat and Power; EQ, Ecosystem Quality; FAO, Food and Agriculture Organization; FE, Freshwater Eutrophication; FET, Freshwater Ecotoxicity; FRS, Fossil Resource Scarcity; GW, Global warning; HH, Human Health; LCA, Life Cycle Assessment; ME, Marine Eutrophication; MET, Marine Ecotoxicity; RS, Resource Scarcity; SOD, Stratospheric Ozone Depletion; SS, Subsystems; TA, Terrestrial Acidification; TET, Terrestrial Ecotoxicity.

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polymer of glucose linked by β-1, 4-glycosidic bonds, is the dominant component among the three fractions (Danmaliki et al., 2016). Cellulose and hemicelluloses are tightly bound to lignin by hydrogen and covalent bonds, forming an integrated complex structure, which limits the yield of bioethanol production (Mukherjee et al., 2016). In order to break the lignin-cellulose-hemicellulose complex, the lignocellulosic biomass must undergo a pretreatment step, either physical, chemical, biological or a mixture of both, making the biomass more accessible for hydrolysis (De Souza et al., 2019).

According to De Souza et al., (2019) (De Souza et al., 2019), the potential of vegetable wastes such as bagasse from sugar cane or sugar beet to obtain bioethanol from their fermentation has already been evaluated. The experience gained in the production of bioethanol from sugarcane bagasse opens the door to the development of similar processes based on other types of waste. Fruit and vegetable residues are rich in sugars, cellulose and hemicellulose with low lignin content (Jahid et al., 2018). In particular, banana waste has ideal characteristics in terms of relative percentage of cellulose and lignin (Guo et al., 2018; Oberoi et al., 2011). According to FAO (Food and Agriculture Organization United Nations, 2019), 622 thousand tons of bananas were produced in Europe in 2018, 62% of the total corresponds to Spain.

During banana cultivation and harvesting, a large amount of waste (leaves, pseudo-stem, rhizome and peel, among others) is collected along with the whole fruit itself at the storage stage when the product does not meet the requirements for sale (e.g., too small size or poor condition of the banana) (Kamal, 2015). Its main components are cellulose (29%), hemicellulose (25%), lignin (10%), fructose (2.7%), glucose (3.2%) and sucrose (7.8%) on a dry basis (Gabhane et al., 2014). However, this composition is subject to banana variety or growing conditions, among others. Currently, banana residues are mainly used to produce animal feed. However, due to its high-water content, additional processing is required to reduce moisture (Padam et al., 2014).

Considering the high amount of water and organic matter present in banana waste, it is not allowed to landfill it according to current legislation (Directive (EU) 2018/850) (European Union, 2018). Approximately, one hectare of banana cultivation produces approximately 220 tonnes of waste per year (Ingale et al., 2014). In this sense, the valorisation of banana peel into bioethanol from a biorefinery approach emerges as an option to be considered in the energy integration of this type of industry.

The simplified bioethanol production process is as follows: after drying and crushing the raw material, the biomass undergoes a hydrolysis stage to saccharify the lignocellulosic material into fermentable sugars, which are used as a growth substrate, producing ethanol and carbon dioxide. At the end of the conversion process, due to the high water content, the ethanol is distilled and dehydrated to obtain concentrated alcohol (De Souza et al., 2019; Tursi, 2019). In addition, a new solid waste stream is generated during the stages that make up the biorefinery, which can be converted into new products such as energy and biofertilisers through the processes involved in anaerobic digestion (Heimersson et al., 2017; Tursi, 2019).

This type of lignocellulosic biomass must undergo pretreatment in order to be valorised into high value-added products. One of the objectives of this pretreatment is to increase the available surface area of the cellulose and to remove part of the lignin and hemicellulose content, which is equivalent to an increase in the efficiency of cellulose hydrolysis (Tursi, 2019). Acid pretreatment is the most promising method for industrial application (Jönsson and Martín, 2016). However, this pretreatment leads to the production of by-products: furfural, 5-hydroxymethyl-2-furfural and carboxylic acids that inhibit enzymatic hydrolysis and fermentation (Guo et al., 2018). In this regard, organic acid pretreatment has emerged as a great potential method in the saccharification of lignocellulosic biomass due to its less hazardous properties as well as the decreased production of fermentation-inhibiting by-products (Rattanaporn et al., 2018; Sahu and Pramanik, 2018). In recent years, the use of different organic acids during pretreatment has been studied in different lignocellulosic biomass for fermentation to obtain bioethanol. A summary of some of the recent studies found in the literature covering this topic is given in Table 1.

Therefore, this study proposes the use of different organic acids in addition to the conventional one (sulphuric) in the acid pretreatment step to reduce the inhibitory effects. To assess the environmental profile of the production process, the overall impacts must be quantified in a number of environmental categories. It should be borne in mind that although bioethanol entails the term "*bio*", it is not synonymous with "*eco*", so it cannot be assumed to have a lower environmental burden than fossil ethanol without a detailed environmental study (González-García et al., 2018). Several environmental analysis methods appear in the literature, such as material flow analysis, energy analysis or life cycle assessment (LCA) (Vandermeersch et al., 2014). In the context of agricultural waste, the literature shows several works implementing the LCA methodology in the framework of the valorisation of this type of fractions.

Therefore, the aim of this study is to analyse different strategies for the valorisation of banana peel waste to obtain bioethanol under an environmental approach to identify the scenario with the lowest environmental burdens. Different organic chemicals will be compared with conventional sulphuric acid because it is the most used solvent in acid pretreatment. The secondary flows originated will be managed in an anaerobic digestion plant, to produce electrical and thermal energy, and achieve a better "closed cycle" approach.

2. Materials and methods

2.1. Life cycle assessment methodology

This study shows the simulation of industrial scenarios based on the valorisation of banana waste and their subsequent environmental analysis, which will allow predicting the environmental implications of each of the processes carried out in the scenarios modelled. Since the LCA methodology makes it possible to evaluate the environmental loads associated with a system, identifying its material and energy flows with the aim of proposing environmental improvement strategies (ISO 14040, 2006), it was the methodology selected for assessment. According to ISO 14040 (ISO 14040, 2006) and ISO 14044 (ISO 14044, 2006), this methodology consists of four phases; goal and scope definition, Life Cycle Inventory, Life Cycle Impact Assessment and results interpretation.

2.2. Goal, scope and system boundaries

The objective of this study is to evaluate the environmental profile of four different banana waste valorisation routes to obtain bioethanol, taking into account that it will be necessary to consider full-scale processes through a process modelling methodology using Aspen Plus® software (Aspentech, 2020). In this research work, the LCA methodology was implemented in a cradle-to-gate perspective, using banana peel as

Table 1
Literature on recent work aimed at the optimisation of acid pretreatment by means of organic acids in lignocellulosic biomass.

Biomass	Organic acid	Reference
Wheat straw	Oxalic, maleic and succinic acid	(Barisik et al., 2016)
Cotton gin	Oxalic, citric, lactic and maleic acid	(Sahu and Pramanik, 2018)
Oil palm trunk	Oxalic, citric and acetic	(Rattanaporn et al., 2018)
Cassava stem	Oxalic acid	(Sivamani and Baskar, 2018)
Corn cob	Oxalic, citric, malic, maleic, malonic, succinic and tartaric acid	(Qiao et al., 2019)

the main raw material. As attribution approach was considered and thus, the environmental impacts associated to the production of bioethanol were quantified from the production of raw materials to the gate of the bioethanol production plant. As it is described below, the scope includes an assessment of the agricultural activities involved in banana production, the bioethanol production as well as the treatment of waste produced in the system.

The scope was not extended to grave because bioethanol is a direct substitute of fossil ethanol and thus, the use and end of life phases are likely to be the same, resulting, in a comparative assessment, in the same relative differences. In an LCA study, the functional unit is the basis for all calculations and the way to express environmental impacts (Ahlgren et al., 2013). Biorefineries fundamentally produce a target product, but in many cases, they produce co-products or by-products (Ahlgren et al., 2013; Ekvall, 2020). For biorefineries, the choice of the functional unit is an important stage since, due to the possible multi-product production of a biorefinery, it can be difficult to identify the main function of the system, which requires additional thought when choosing the functional unit (Ahlgren et al., 2013). There are three different ways in the choice of the functional unit: by quantity of input (raw material), by the output of a single product or the combination of several output products (Ahlgren et al., 2013).

Consequently, bearing in mind that the main objective of the system is the valorisation of banana peel wastes into bioethanol, the selection of a reference unit based on the target product guarantees consistency throughout the study as well as facilitates comparison with other studies and alternatives. Accordingly, one kilogram of bioethanol was selected as functional unit, being consistent with other studies available in the literature (Ekman and Börjesson, 2011; Gullón et al., 2018; Piemonte, 2012). As mentioned above, three different organic acids were used in the acid pretreatment step, the chosen ones being tartaric, oxalic and citric acids. These acids were taken into account based on the study by Guo et al. (2018) (Guo et al., 2018), which focused on the optimisation of acid pretreatment for bioethanol production using banana peels as lignocellulosic biomass.

In that study, the aim was also to know the production yield when using each of the organic acids in addition to the conventional sulphuric acid. It was concluded that tartaric acid yielded the highest amount of ethanol, followed by oxalic and sulphuric acid and finally citric acid. In this LCA study, the environmental impacts associated with the construction and installation of the biorefinery plant during its lifetime were assumed non-significant, so no infrastructure process was considered in line with other similar studies (Jeswani et al., 2015; Karlsson et al., 2014). Finally, the transport of the main raw material (i.e., banana waste) and chemicals to the biorefinery plant as well as solid waste from the valorisation process to the treatment stage was considered within the system boundaries.

Distribution distances of 100 km and 30 km were assumed up to the biorefinery and digestion plant gates, respectively. These assumptions are consistent with the literature (Hajjaji et al., 2013; Lijó et al., 2017). In addition, the transport of chemical products, with an average distance of 600 km, has been considered (Pérez-López et al., 2014). Fig. 1 describes the system boundaries under consideration, the same in all scenarios proposed for analysis. All scenarios are composed of six sub-

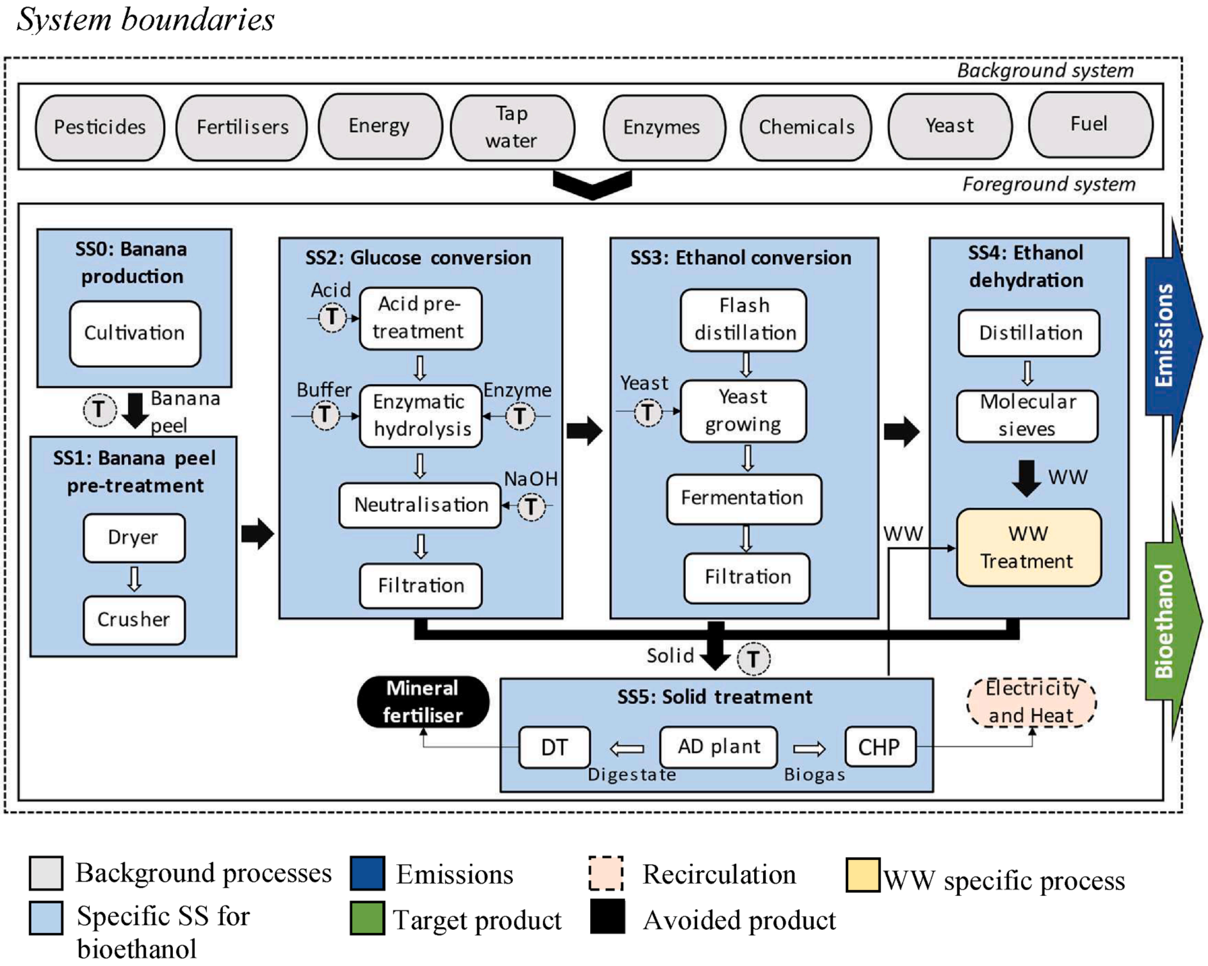

Fig. 1. System boundaries of the scenario proposed as a biorefinery strategy. Acronyms: AD – Anaerobic Digestion, CHP – Combined Heat and Power, DT – Digestate Treatment, SS – Subsystem, T – Transport, WW – Wastewater.

systems (hereafter SS); banana production (SS0), banana peel pretreatment (SS1), glucose conversion (SS2), ethanol conversion (SS3), ethanol dehydration (SS4) and solid treatment (SS5), with SS0 and SS1 being common to all scenarios studied.

2.3. Description of the full-scale production facilities

Process modelling allows, in addition to the design and optimization of production processes, the identification of inventory data to carry out the environmental assessment (Morales-Mendoza et al., 2018). Drawing on published studies on banana peel valorisation into bioethanol (Danmaliki et al., 2016; De Souza et al., 2019; Gebresemati and Gebregergs, 2015), four different scenarios (scenarios A-D) based on the use of different types of organic acids can be envisaged. Figure S1 shows the flowchart corresponding to the different valorisation sequences proposed.

Subsystem 0 (banana production, SS0) is the stage where banana is cultivated producing the banana fruit, the main product, and banana wastes being the latter collected during banana fruit harvesting. This stage includes all the activities performed in the field from soil management to harvesting. Information concerning the requirement of the different inputs such as diesel, agrochemicals and machinery as well as on-field emissions derived from the agrochemicals application was taken from Velásquez-Arredondo et al. (2010). As mentioned above, one hectare of banana cultivation produces approximately 220 tons of waste per year (Ingale et al., 2014) mainly constituted by peels, leaves and pseudo-stems (Kamal, 2015).

It was assumed that the banana waste that reaches the biorefinery gate has a composition analogous to the banana peel. Once the waste reaches the plant, it will be stored until it is led to the pretreatment stage. The inventory data reported by Velásquez-Arredondo et al. (2010) have been taken into consideration. Bearing in mind that approximately 25% of the banana fruit produced is peel (Velásquez-Arredondo et al., 2010), a mass allocation has been considered to allocate the burdens between both co-products (banana fruit and peels) derived from the cultivation stage (SS0).

Subsystem 1 (banana peel pretreatment, SS1) starts with drying the banana peels at 60 °C to achieve a moisture content of 10% from a feedstock with an initial moisture content of 89% (Guo et al., 2018). They are then taken to a shredding unit to reduce the particle size to 5 cm. The next stage corresponds to Subsystem 2 (glucose conversion, SS2) starts with acid pretreatment. As mentioned above, four different acids will be used in different scenarios. Therefore, sulfuric acid (the conventional one) corresponds to Scenario A and tartaric, oxalic and citric acids are considered in Scenarios B, C and D, respectively. The acid pretreatment process is carried out for 1 h in a stirred tank at 80 °C with a solid:liquid ratio of 1:15 (w/w) and acid concentration of 0.2%.

This acid condition applies for each of the scenarios. Enzymatic hydrolysis is the step after acid pretreatment. Previously, cellulase enzyme solution is prepared in 0.1 M citrate buffer solution and added to the reaction mixture for 1 h at 50 °C for conversion of cellulose to glucose which must be neutralised with 1 M NaOH before fermentation. In addition, the mixture is filtered to separate the non-soluble solid stream from the neutralised syrup. The yeast fermentation process is conducted in Subsystem 3 (ethanol production, SS3). This process takes place at 30 °C for 36 h (Guo et al., 2018). Once the fermentation is over, carbon dioxide, produced during this process, is discharged as exhausted gas and the suspension is filtered to obtain a new residual solid stream while the rest is sent to Subsystem 4 (ethanol dehydration, SS4) to be subjected to a double distillation to concentrate and recover as much ethanol as possible in SS4.

Concerning the stream entering the SS4, it is preheated before being introduced into the first distillation column. Finished the distillation, ethanol is dehydrated using molecular sieves to produce anhydrous ethanol (99.8%) (Lauzurique-Guerra et al., 2017), the target product. An important amount of wastewater is obtained from the dehydration which is treated in a wastewater treatment plant. The wastewater treatment process, according to the Ecoinvent® database (Moreno Ruiz et al., 2018), consists of three stages; mechanical, biological and chemical. Treated water is finally discharged in the environment (0.9 m^3 per m^3 of wastewater) (Moreno Ruiz et al., 2018).

In Subsystem 5 (solid treatment, SS5) the solid outflows from SS2, SS3 and SS4 (see Fig. 1) are digested under mesophilic conditions (≈35 °C) (Achinas et al., 2019) to obtain two co-products: biogas and digestate. The biogas is burned in a cogeneration unit (CHP, Combined Heat and Power) (Heimersson et al., 2017) to be transformed into electricity and heat. A CHP unit with electrical and thermal efficiencies of 32% and 50%, respectively (Arias et al., 2020a), was considered. Electricity and heat are recycled into the biorefinery plant to satisfy energy requirements. The digestate obtained could be used in farming activities as biofertiliser after suffering a specific treatment to meet the requirements according to legislation (Heimersson et al., 2017). Therefore, the digestate is pasteurised (70 °C for 1 h) and dewatered using a centrifuge (Banks et al., 2011). The liquid fraction obtained is sent to the wastewater treatment plant. The biofertiliser pasteurised is stored until its use as fertiliser on the crop (Holm-Nielsen et al., 2009). The amount of biofertiliser produced was managed as an avoided product taking into account the nutrients content of the digestate (Ascher et al., 2020).

2.4. Life cycle inventory

This stage consists of summarising the input and output data of the system under consideration for the environmental assessment. In this study, inventory data, such as energy requirements (electrical and thermal) of all equipment, chemical consumption and process water have been obtained from process simulations using Aspen Plus® software, taking as input data the experiments conducted at laboratory scale developed by Guo et al. (2018) and Lauzurique-Guerra et al. (2017). The mass and energy balances resulting from process simulation are used as the basis for the life cycle inventory (see Table 2) of each scenario. The treatment capacity for the scenarios is 300 kg of banana peels per batch at approximately 90% moisture.

When conducting the environmental analysis, the heat source is obtained from steam derived from the biorefinery, while the cooling energy consists of the recovery of the refrigeration utility in a cogeneration unit. Inventory data associated with their production was taken from the Ecoinvent® database (Moreno Ruiz et al., 2018) but updated considering the Spanish electricity mix for 2019 (Spanish Electrical Network, 2020). Inventory data for SS0, belonging to banana cultivation, was obtained from (Velásquez-Arredondo et al. (2010). Concerning the anaerobic digestion (SS5) and wastewater treatment (SS4) processes, inventory data were also taken from the Ecoinvent® database.

As indicated above, the biogas obtained is converted into electricity and thermal energy in a CHP unit to be used as renewable energy in the plant (Arias et al. 2020a). To calculate the amount of biogas generated and the energy produced from the biogas burned, information and the equations from Arias et al. (2020a) were taken, although adapted to the characteristics of the stream, as describe below:

$$V_{Biogas} = \eta \cdot m_{VS} \cdot m_{peel} \tag{1}$$

$$Energy\ production_{e/t} = V_{Biogas} \cdot \rho \cdot LHV \cdot N_{e/t} \tag{2}$$

where;

V_{Biogas}: Volume of biogas generated (per m^3 CH_4).

η: Methane yield per volatile solid (VS) = 0.227 m^3 $CH_4 \cdot kg^{-1}$ VS (Zheng et al., 2013).

m_{VS}: Mass of volatile solids present in banana peel (0.069 kg VS· kg^{-1} banana peel) (Achinas et al., 2019).

m_{peel}: Mass of banana peel treated by AD (kg banana peel)

ρ: Methane density (0.656 $kg \cdot m^{-3}$ CH_4).

Table 2

Life cycle inventory for the valorisation of banana peel from all the proposed scenarios. Scenario A - Sulphuric acid based; Scenario B - Tartaric acid based; Scenario C - Oxalic acid based; Scenario D - Citric acid based.

Inputs from Technosphere		Scenario A	Scenario B	Scenario C	Scenario D
SS0: Banana production					
	Urea (kg)	57.8			
	Potassium chloride (kg)	78.6			
	Nitrogen fertiliser (kg)	2.0			
	Phosphate fertiliser (kg)	2.2			
	Ammonium sulphate (kg)	6.9			
	Pesticide (kg)	0.92			
	Diesel (kg)	12.4			
SS1: Banana peel pretreatment					
	Banana peel (kg)	300.0			
	Air (kg)	2453.0			
	Electricity (kWh)	185.5			
	Heat energy (kWh)	23.9			
	Transport (km)	100.0			
SS2: Glucose conversion					
	Acid (kg)	1.79	1.77	1.24	1.11
	Sodium hydroxide (kg)	1.32	2.53	1.09	11.7
	Tap water (kg)	125.9	134.0	120.9	142.9
	Enzyme (g)	6.2			
	Buffer solution (kg)	5.0			
	Yeast (g)	129.4	129.4	117.2	159.2
	Electricity (kWh)	1.4	2.0	1.8	1.4
	Heat energy (kWh)	32.4			
	Cooling energy (kWh)	33.6			
	Transport (km)	600.0			
SS3: Ethanol conversion					
	Electricity (kWh)	0.25	0.28	0.28	0.21
	Heat energy (kWh)	281.5	298.8	267.8	288.1
	Cooling energy (kWh)	281.5	298.8	267.8	288.1
SS4: Ethanol dehydration					
	Electricity (kWh)	0.24	0.71	0.69	0.54
	Heat energy (kWh)	21.3	22.3	21.0	30.2
	Cooling energy (kWh)	19.9	20.8	19.7	28.0
	Wastewater treatment				
	Electricity (Wh)	73.0	90.2	88.0	40.8
	Heat energy (Wh)	5.6	6.9	6.8	3.1
	Chemicals (g)	10.5	13.0	12.7	5.9
SS5: Solid treatment					
	Tap water (kg)	24.6	25.9	23.7	28.1
	Electricity (Wh)	785.9	828.1	755.4	896.2
	Heat energy (kWh)	16.1	17.0	15.5	18.4
	Transport (km)	30.0			
Outputs to Technosphere		Scenario A	Scenario B	Scenario C	Scenario D
Product					
	Bioethanol (kg)	1.93	2.19	2.13	1.84
Emissions					
	To air				
	Steam (from SS1) (kg)	2717			
	CO_2 (from SS3) (kg)	2.1	2.4	2.3	1.9
	CO_2 (from SS5) (kg)	32.3	34	31.1	36.8
	N_2O (from SS5) (g)	5.1	5.3	4.9	5.8
	H_2S (from SS5) (g)	13.8	14.5	13.3	15.7
	CH_4 (from SS5) (g)	155.3	163.7	149.4	177.2
	To water				
	NH_4^+ (from SS5) (mg)	14.3	15.0	13.7	16.3
	NO_3^- (from SS5) (mg)	456.8	481.4	439.3	520.9
	NO_2^- (from SS5) (mg)	14.3	15.0	13.7	16.3
	N (from SS5) (mg)	16.8	17.7	16.1	19.1
	P (from SS5) (mg)	10.8	11.4	10.4	12.3

LHV: Lower Heating calorific Value of methane (50 MJ·kg^{-1} CH_4).

$N_{e/t}$: Energy efficiency of the cogeneration unit (N_e=32%; N_t=50%)

The amount of energy produced per scenario is shown in Table S1. This energy is considered as an output from SS5, but not from the system as it is recirculated into the biorefinery. It was assumed that the electricity and heat produced may substitute an equivalent amount of energy from the energy profile of the biorefinery plant. Therefore, the avoided energy production was included within the system boundaries (see Fig. 1). Table S1 shows a summary of the waste streams brought to treatment (anaerobic digestion or wastewater treatment) as well as the amounts of energy and mineral biofertilisers obtained. Finally, Table S2 summarises the detailed information of the data sources for the background processes.

As indicated above, it has been assumed that the digestate obtained from anaerobic digestion could be used as biofertiliser in agricultural activities. To do so, it was considered the amount of mineral fertilisers that could be substituted by the digestate, taking into account mineral substitution ratios identified in the literature (Ashekuzzaman et al., 2021). Therefore, each kilogram of N, P and K contained in the digestate is equivalent to 0.52, 0.95 and 1 kg of mineral N, P and K, respectively. It should be noted that the main product of the biorefinery plant is bioethanol. The energy obtained in the CHP unit has been subtracted from the total amount of energy required in the scenarios. In addition, the use of digestate from anaerobic digestion has also been considered and can be used as a fertiliser for crop fields. Thus, in this study a system expansion strategy is realised to avoid an allocation of environmental burdens. It is also worth mentioning that the environmental impacts are expressed per kg of main product, that is, the bioethanol

2.5. Life cycle impact assessment methodology

For the environmental analysis, two impact assessment methods were considered. Firstly, the hierarchical midpoint method of ReCiPe 2016 V1.04 World (2010) (Huijbregts et al., 2016) was used to perform the environmental profiling. Thus, the characterisation factors reported by this method were considered to estimate the environmental burdens in terms of the following impact categories: global warming (GW), stratospheric ozone depletion (SOD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and fossil resource scarcity (FRS). On the other hand, the hierarchical Endpoint method of ReCiPe 2016 V1.04 World (2010) H/H (Huijbregts et al., 2016) was considered to compare the profiles of bioethanol with each other. With this endpoint approach it is possible to have a single score of the general impacts, allowing a better dissemination of the environmental results to the stakeholders (Arias et al., 2020b).

Three endpoint categories, expressed in millipoints – mPt, were studied to obtain a direct comparison. Human Health (HH), Ecosystem Quality (EQ) and Resource Scarcity (RS) were these endpoint categories. All previously assessed midpoint categories contribute to the endpoint results, resulting in a single environmental score estimate. Therefore, the corresponding normalisation and weighting factors established by the endpoint method were considered (Huijbregts et al., 2016). These normalization factors, defined in Table S3, were taken from the ReCiPe 2016 Endpoint method. Furthermore, Figure S2 shows an explanatory diagram of the endpoint method carried out. Finally, it should be added that the computational implementation of the life cycle inventory data was carried out using the SimaPro v9.0.0 software (Consultants, 2019).

3. Results

3.1. Global environmental results

Table 3 shows the results of the environmental assessment for the most relevant impact categories studied by the hierarchical midpoint method ReCiPe 2016 V1.04. Bearing in mind these results, notable differences in the bioethanol production profiles are appreciated depending on the chemical used in the glucose conversion stage (SS2). Consequently, Scenario D (based on the use of citric acid) is the one that presents the worst environmental profile since the environmental burdens, for all impact categories, are higher than those from the other scenarios. Scenario A (based on sulphuric acid), has the second worst environmental profile, with decreases of between 10% and 27% depending on the impact category with respect to the burdens of Scenario D.

On the other hand, Scenario C (based on oxalic acid) reports the lowest scores in all impact categories except for the SOD and TET, with Scenario B (based on tartaric acid) having the lowest impact for these categories. Compared to Scenario D, the environmental burdens of Scenario C decrease by 22% to 34% depending on the impact category. Besides, special attention should be paid to the fact that, when performing the environmental analysis, the same organic chemical compound (see Table S2) is used for both tartaric and oxalic acid due to the lack of specific inventory data for their production. Therefore, the production of an unspecified organic chemical was considered. Furthermore, the production yields are similar and there is a small difference between the amounts of inputs and outputs between the two valorisation scenarios, being higher for Scenario B (see Table 2).

To justify the environmental results of the different profiles evaluated, an analysis of the contribution by subsystems of environmental burdens per scenario in each of the impact categories studied was carried out, with the aim of identifying the environmental hotspots.

3.2. Identification of hotspots

The identification of environmental hotspots emerges, in this study, to know the reason of the main environmental impacts and as a key issue to propose new actions to improve the different modeled valorization schemes. For this reason, the operations involved in each of the banana waste valorisation systems responsible for the greatest environmental burdens were identified and discussed. The relative contributions of environmental impacts per subsystems involved in the different valorisation scenarios are detailed in Fig. 2. Firstly, it can be seen that the contributions in all scenarios are practically identical, differing in some respects.

According to the environmental results of Scenario A (sulphuric acid), SS0 (banana production) contributes mainly in TET, representing 58% of the total burdens. In the SOD and TA, this subsystem contributes approximately 38% each. The rationale behind these results is due to the requirement of mineral fertilisers in agricultural activities and the corresponding production and derived on-field emissions. Contributions from SS1 (banana peel pretreatment) to TA, FE, FET and MET range between 38% and 48% of the total contributions to these categories. The reason behind these results is due to the production of the electricity required to reduce both the moisture content and the size of the residues. According to the results, SS2 (Glucose conversion), despite being the subsystem where chemicals intervene in the biorefinery plant, does not

Table 3
Environmental characterisation results per impact category and functional unit (1 kg of bioethanol) for each scenario under study. Scenario A – Sulphuric acid; Scenario B – Tartaric acid; Scenario C – Oxalic acid; Scenario D – Citric acid.

Impact category	Unit	Scenario A	Scenario B	Scenario C	Scenario D
GW	kg CO_2 eq	219.9	203.3	194.9	250.5
SOD	mg CFC11 eq	61.7	53.8	57.3	73.4
TA	g SO_2 eq	455.5	406.6	406.2	519.2
FE	g P eq	22.5	20.9	20.4	28.3
ME	g N eq	4.58	4.32	4.30	5.70
TET	kg 1,4-DCB	189.3	168.7	170.3	226.3
FET	g 1,4-DCB	930.9	853.1	845.1	1276.5
MET	kg 1,4-DCB	1.33	1.21	1.20	1.79
FRS	kg oil eq	65.9	61.3	58.7	73.6

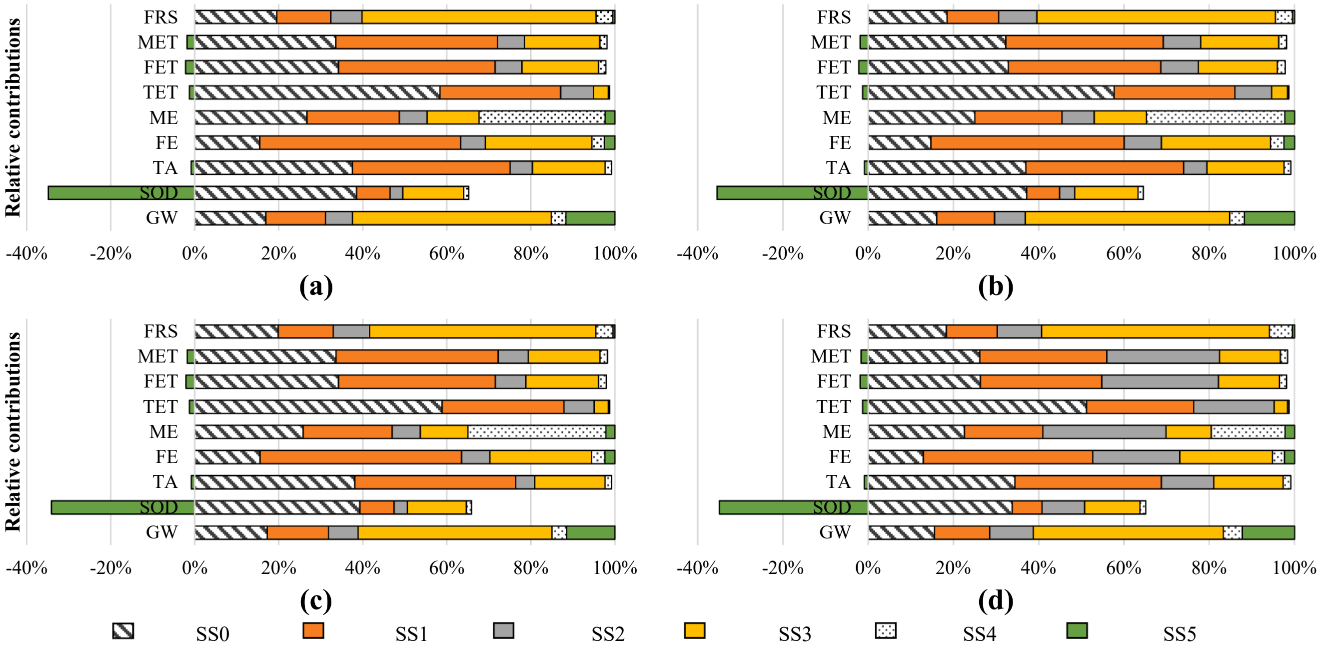

Fig. 2. Relative contributions of environmental impacts per SS involved in the valorisation scenarios (functional unit: 1 kg bioethanol). (a) Scenario A - Sulphuric acid based; (b) Scenario B - Tartaric acid based; (c) Scenario C - Oxalic acid based; (b) Scenario D - Citric acid based.

report an outstanding contribution to the global profile because of the small amount of chemicals needed at this stage. Moreover, the energy requirements in this subsystem are not very high compared to the rest of the subsystems (see Table 2).

SS3 (ethanol conversion) contributes significantly to GW and FRS, representing 47% and 56% of the total, respectively. This is due to the large amount of thermal energy required by the units that involved this subsystem. Specifically, flash distiller, condenser, fermenter and filtration unit require 75% of heating energy and 85% of cooling energy over the total required in the entire plant (see Table 2). SS4 (ethanol dehydration) reports negligible contributions to all impact categories (below 5% of the total), except for ME, with a contribution of 30% of the total. The reason behind these results is the background processes involved in wastewater treatment. As mentioned above, the use of digestate as an organic fertiliser implies the reduction of mineral fertilisation according to the approach considered in this study with the aim of avoiding the burdens allocation. Therefore, a negative contribution (environmental credit) of SS5 (solids treatment) can be identified in SOD, with a ratio of 35% of the total, and to a lesser extent (below 2%) in TA, TET, FET and MET. This is due to the avoided mineral fertilisers production because of the digestate production.

Fig. 2b shows the relative contributions of the environmental impacts involved in Scenario B (based on tartaric acid). It can be observed that the profile is close similar to Scenario A detailed in Fig. 2a but with some specific differences. It should be noted that SS0 contributes to TET (57% of the total) and SOD and TAT (37% each of the total). The reason behind these scores is associated with the production of the fertilisers required together with the on-field emissions derived from their application. Contributions from SS1 to TA, FE, FET and MET range between 36% and 45% of the total contributions to these categories. Electricity required to pretreat the banana peels (moisture removal and size reduction) is the main reason for these results.

Once again, SS2 highlights for its small contribution to the overall environmental profile (below 10% of the total), due to the small amount of energy and chemicals needed. In the same way as Scenario A, SS3 contributes to GW and FRS notoriously (48% and 56% of the total, respectively, due to the production of the thermal energy required by the units that make up SS3. Furthermore, according to the results, SS4 contributes to MET (about 32% of the total) due to the processes involved in wastewater treatment. Finally, SS5 has negative values in SOD, TA, TET and FET and MET, with the most notable value in SOD, contributing 36% of the total. As in the sulphuric acid-based scenario, these environmental credits are due to the avoided production of mineral fertilisers.

Fig. 2c displays the relative contributions of the environmental impacts involved in the oxalic acid-based scenario (Scenario C). As in the previous scenarios, the contributions to the environmental profile follow the same trend. The contributions of SOD, TA and TET to SS0 are 39%, 38% and 59% respectively. The reasons for these contributions are the production of the fertilisers and the emissions from their application. The contributions of SS1 to TA, FE, FET and MET range between 38% and 42% of the total contributions to these categories. Again, the main reason for the results is the energy required to carry out the pretreatment of banana peels.

Contributions from the glucose-conversion subsystem (SS2) to the environmental profile remains low (below 10% of the total), due to the small amount of chemicals required, in addition to the low energy requirements compared to the other subsystems. The contribution to GW and FRS, significantly, by SS3 is 46% and 54% of the total, respectively, due to the high energy requirements of the equipment that constitute this subsystem, especially the flash distiller, condenser and fermenter. As in the previous scenarios, the contributions of SS4 to the impact categories are negligible, apart from ME (30% of the total), due to the processes involved in wastewater treatment. Lastly, the contributions of SS5 to the environmental profile show negative values in, the most remarkable value being that of SOD (34% of the total), due to the avoided production of mineral fertilisers.

To conclude the overall analysis of the environmental profiles, the relative contributions of the environmental impacts surrounding Scenario D, based on citric acid, are presented in Fig. 2d. From the results, SS0 constitutes a critical point, from the environmental point of view, with contributions for TET with 51% of the total and also for SOD and TA with 34% of the total. The reasons are the same as for the other scenarios, the production of fertilisers and the emissions from their use on the fields. The contributions to TA, FE, FET and MET by SS1 (banana peel pretreatment) range between 28% and 40% of these categories. The energy to remove moisture from the peels as well as to carry out the shredding are the main reason for these results.

In contrast to the other scenarios, SS2 has a more notable contribution for some of the impact categories such as FE, ME, TET, FET and MET, with contributions ranging between 20% and 30% of the total. The reason for this variation will be analysed later since no cause is observed to justify these first-hand results. SS3 operations, where glucose is converted into ethanol, are a hotspot for GW and FRS, with contributions of 45% and 54% of the total, respectively. The thermal energy required by the equipment in this subsystem (mainly, flash distiller, condenser, fermenter and filtration unit) is the main cause of the environmental hotspot. Finally, there is a decrease in the contribution of SS4 in ME compared to the other scenarios (around 15%). Moreover, the environmental credit in SOD for SS5 follows the same trend as in the other scenarios, being around 35%. As indicated above, the avoided production of mineral fertilisers is the main cause of this negative value.

3.2.1. *Detailed environmental analysis of subsystem 2 (glucose conversion)*

As shown in Fig. 2 above, there are some small differences between the environmental profile of Scenarios A, B and C with respect to Scenario D, and this difference is reflected in SS2. Scenarios A and B require the largest amount of acid, with Scenario D requiring the least amount. In addition, the amount of sodium hydroxide is much higher in Scenario D than in the other scenarios, requiring between 78 and 90% more. It should be added that Scenario C requires the least amount of sodium hydroxide of the four scenarios.

Since SS2 is the most notable cause of the variation between the different environmental profiles analysed for the different valorisation scenarios, it is necessary to carry out an in-depth study of this subsystem to identify the main hotspot. This variation mentioned is due to an increase in the values of the characterisation results, but the values for the rest of the subsystems do not undergo a notable change as they do in SS2. The environmental results corresponding to SS2 of the different valorisation scenarios are shown in Fig. 3. Considering the environmental profile of the sulphuric acid-based scenario (Scenario A) (see Fig. 3a), the production of cooling energy requirements can be considered as the environmental hotspot, with contributing ratios ranging between 33% and 65% for all impact categories, except for ME and TET.

On the other hand, the production of the buffer solution involves also outstanding contributions in categories such as FET and MET, being the hotspot in ME and TET. In addition, the background processes involved in the production of the acid compound are the environmental hotspots in TA and TET (31% of the total). Finally, the background processes involved in the production of enzymes is the hotspot in ME, with a contribution of 27% of the total.

Fig. 3b details the distributions of burdens derived from SS2 in Scenario B (based on tartaric acid). As in the previous profile, the production of the cooling energy required is the main hotspot, contributing for all impact categories, except for ME and TET, with values ranging from 23% to 55%. As in the previous case, buffer solution production contributes notoriously to environmental burdens for MET and TET, besides being the hotspot in ME and FET categories. The background processes involved in the acid production presents highlighted contributions to TA, FET, MET y FRS, being an environmental hotspot in TET. Moreover, the background processes involved in the production of sodium hydroxide play a key role in FE y TET (around 35% of the total for each one) and their implications are prominent in SOD, TA, ME, FET and MET. Finally, the production of the buffer solution involves also outstanding contributions in categories such as FET and MET, being the hotspot in ME and TET. To end, the background processes involved in the enzymes production are the hotspot in ME (22% of the total).

Next, Fig. 3c details the profile from SS2 in Scenario C. As in the previous profiles, the results again show that the production of cooling energy requirement is the main environmental hotspot for all impact categories (except for ME and TET), with values ranging from 29% to 43%. Again, the buffer solution production involves remarkable contributions in TET and MET, being the environmental hotspot in ME and FET. Furthermore, the background processes involved in the production of the acid compound constitute the environmental hotspot in TET (25% of the total). The hotspot in ME (26% of the total) is due to the background processes involved in the enzymes production. Lastly, the buffer solution production involves also outstanding contributions in categories such as FET and MET, being the hotspot in ME and TET.

Finally, Scenario D reports a shift in the profile of SS2 (see Fig. 3d) as

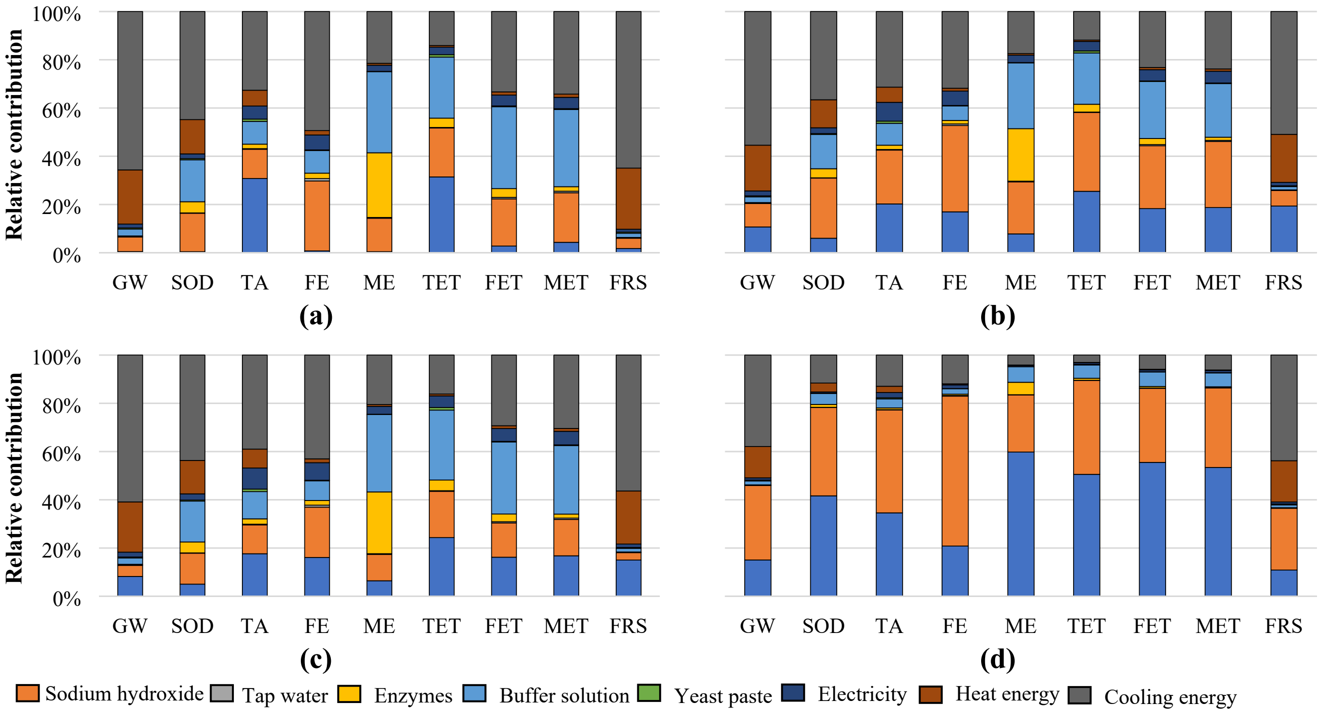

Fig. 3. Distribution of burdens between contributing inputs/outputs involved in Subsystem 2 (glucose conversion); (a) Scenario A – Sulphuric acid based; (b) Scenario B - Tartaric acid based; (c) Scenario C - Oxalic acid based; (d) Scenario D - Citric acid based.

difference to the previous scenarios. The background processes involved in the production of acid are the environmental hotspot in ME, TET, FET and MET, with a range of values between 51% and 60%. On the other hand, the main hotspots in TA (43% of the total) and FE (62% of the total) are associated with the background processes involved in sodium hydroxide production. As a final point, the production of the cooling energy requirements can be considered as the environmental hotspot in GW and FRS because of their dependence on fossil sources, with ratios of 38% and 44% of the total, respectively.

3.3. Environmental impact assessment though the endpoint perspective

As indicated in Section 2.5, to obtain a more accurate information on which of the four valorisation scenarios studied has the best environmental profile, an environmental comparison was performed based on an endpoint method. The result of the comparison is illustrated in Fig. 4. The single score corresponding to Scenario A (3223 mPt), Scenario B (2979 mPt), Scenario C (2859 mPt) and Scenario D (3664 mPt) are in the same order of magnitude although with differences. If a detailed analysis of the scores and the distribution between the level of damage (HH, EQ and RS) is performed, the damage to HH dominates the environmental score (80% of the total) in all scenarios. The main cause of these high contributions is the use of fossil resources in their background systems, specifically in the production of the high energy requirements in the biorefinery plants.

The contribution to the single score from EQ is around 13% for all the valorisation scenarios, which is associated with emissions from the background processes involved in all valorisation scenarios, with a particular focus on those associated with the production of energy requirements. The contribution from RS is of 7% regardless of the scenario and is associated with the use of fossil resources in the background processes implicated in the biorefinery plant. Given the results, it can be concluded that the best valorisation scenario with the best environmental profile is the scenario based on tartaric acid, as it has the lowest environmental score. On the other hand, the scenario based on citric acid reports the highest single score, making it the worst route for the recovery of banana waste to obtain bioethanol from an environmental approach. Furthermore, the low production yield of this scenario must be considered. In this regard, future activities should be focused on optimizing the valorisation routes specifically in terms of energy requirements and bioethanol yields.

3.4. Prospect for improvement

Once the environmental analysis was carried out and the main hotspots were identified, several improvement options were proposed to reduce the environmental burdens of all the proposed valorisation scenarios. The energy required to dry the banana peels in SS1 is responsible for a high environmental burden. Therefore, it is proposed to look for other drying methods that allow a reduction in energy, (e.g., air drying) or less conventional methods such as microwave drying (Khodifad and Dhamsaniya, 2020). It is worth highlighting another possible reduction in environmental loads that could also be a benefit to the production of the biorefinery plant.

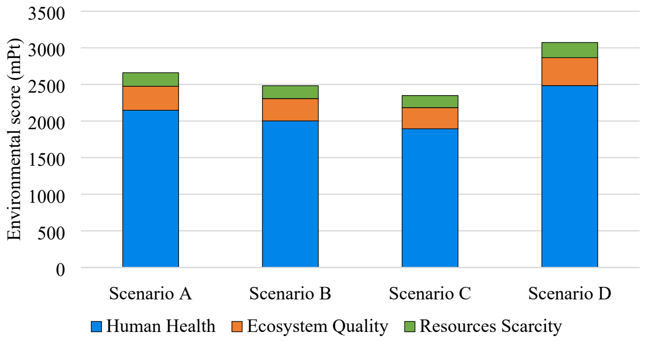

Fig. 4. Comparison of life cycle impacts and damage contributions on Human Health, Ecosystem Quality and Resources Scarcity.

The equipment whose function is to cool the system streams uses cooling energy recovered from the refrigeration utility in a cogeneration unit. However, further research should be focused on an energy study to understand the feasibility of reusing the thermal output streams from the units of the biorefinery plant itself. Nevertheless, this analysis only considers the amount of energy required by the system, without taking into account the quality of the energy, resulting in unreliable information for decision-making related to sustainability in terms of cost, risk, etc. (Aghbashlo et al., 2021). This can be solved by exergy, a concept that considers both the amount and quality of energy, which gives more meaningful results. Exergy analysis identifies the location, magnitude and causes why an energy system is inefficient. Thus, the concept of exergy can be integrated into the LCA methodology, offering a new sustainability assessment tool known as "exergy-environmental analysis" (Aghbashlo et al., 2021). For future research, this new analysis method could be implemented in this biorefinery study.

The enzymatic activity during hydrolysis also becomes a key point for improvement, as a deeper understanding of this process, aimed at depolymerisation of plant biomass, would allow optimisation of its operating conditions. An example of this can be seen in the study by Sepulchro et al. (2021) (Sepulchro et al., 2021), where they use lytic polysaccharide monooxygenase (LPMO) enzymes as an important aid in the enzymatic depolymerisation of lignocellulosic biomass, facilitating its valorisation into high value-added products. Finally, it should be noted that this study proposes the valorisation of waste fractions that cannot be used for food purposes, and gives a sustainable use to agricultural waste, without neglecting that its potential as a source of fermentable sugars.

However, the possibilities of its adoption by the industry depend on the resourcefulness and implementation of the best and most viable pretreatment of biomass adequacy (key point), combined with an efficient fermentation process of fermentable sugars and purification of the final product. In addition, one of the main challenges of using biomass as a feedstock for biofuel production is the high innate variability between different biomass types. This variability, due to different growth and harvesting conditions, raises problems for conversion processes, which often require physically and chemically uniform materials (Williams et al., 2017). Therefore, investment in this type of research is of vital importance not only for scientific progress, but also for economic, environmental, social and human development. On balance, the environmental assessment concludes that the impacts are not severe and are the result of parallel or indirect activities.

4. Conclusions

The comparative environmental analysis based on the LCA methodology of different valorisation scenarios of banana peel waste was carried out to obtain bioethanol as the main high-added value product, under a biorefinery approach. The non-common point among the scenarios proposed was the acid pretreatment stage being the sulphuric acid as the conventional one and the tartaric, oxalic and citric acids the most innovative ones. The use of different pretreatment acids derived into different environmental profiles and consequences. By reporting environmental results, among the valorisation scenarios to obtain bioethanol, it was concluded that oxalic acid-based method is the most environmentally friendly since it has associated the lowest environmental single score. On the contrary, according to this study, the citric acid-based method was the one that contributes most to the environmental impact categories either in terms of midpoint or endpoint methods.

LCA methodology makes it possible to establish a comparative framework that can represent a useful tool for identifying the advantages and disadvantages of each selected technique, as well as the possibility of identifying the operating parameters on which to act in the short and medium term. The subsystems responsible to produce the raw material and its pretreatment were the main responsible for the environmental loads of all the proposed valorisation scenarios. When citric acid is considered, the background process involved in its production was identified as the main cause of environmental loads. In addition, it is not considered a good option not only because of the low performance of bioethanol extraction, but also because it is not environmentally friendly.

Finally, the production of energy requirements is the main hotspot in numerous processes: the drying of the banana peels to reduce the moisture content, the heating of the inlet stream prior to fermentation and the later distillation in the corresponding columns are the processes that required the highest energy demands. Therefore, the use of renewable energy sources to satisfy energy requirements instead of taking it directly from the national grid, the optimisation of energy requirements by reusingsome internal steams or considering alternative drying methods should receive special attention with the aim of improving the environmental profiles of all scenarios. Further, as future work, a sensitivity study should be carried out considering other solid waste treatments that consider energy valorisation, such as pyrolysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2022.02.005.

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