



# Environmental insights of bioethanol production and phenolic compounds extraction from apple pomace-based biorefinery

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## ABSTRACT

Food waste is one of the main challenges of solid waste management throughout the food supply chain. Apples are one of the most widely consumed fruits worldwide, the production of which is accompanied by the generation of pomace as a by-product with valuable nutrients. This research presents the first environmental insights of a multiproduct apple pomace-based biorefinery through a life cycle perspective. The design and process modelling of this platform aims to produce bioethanol and extract total phenolic compounds (TPC) towards an efficient use of the pomace obtained from the apple manufacturing industry (juice production). Bioethanol production consist of pressing, fermentation, distillation as the main processes; while in the case of TPC, two extraction techniques were evaluated: i) solvent extraction (mainly water), and ii) the Soxhlet method. The life cycle analysis followed an attributional *cradle-to-gate* approach considering different midpoint impact categories from the ReCipe 2016 method such as Global Warming (GW), Eutrophication, Human Toxicity, Fossil Scarcity, among others. The results indicate that bioethanol production encompasses a GW profile of 3.17 kg of CO<sub>2</sub> eq per kg of product, being the vinasse treatment (subproduct after distillation) the most impactful stage. Phenolic compounds extraction with water achieves a total value of 5.8 kg CO<sub>2</sub> eq per g of TPC, while 0.22 kg CO<sub>2</sub> eq per g of TPC is obtained with Soxhlet technique. Furthermore, a sensitivity analysis is addressed to improve the environmental profile of bioethanol and TPC. This research demonstrated the relevance of process design on the environmental performance of bioethanol and TPC, where stillage treatment has a key contribution to the results and the use of solvents in TPC extraction, although improving extraction yield, leads to a higher environmental impact.

## 1. Introduction

One of the global environmental challenges facing society is to establish an adequate and effective management of the waste generated, which is very diverse. Municipal solid, industrial hazardous, medical, and agricultural waste are some examples of this variety. Green and food waste is one of the most concerning types since accounts for almost half of the world's total residue generation (Costa et al., 2022). Food waste includes uneaten dumped food and food discarded during production, manufacturing, retailing, and consumption (An et al., 2024). Among these stages of the value chain, manufacturing represents 39 % of the waste produced, which is caused by overproduction or poor appearance

for sale (Santiago et al., 2021).

To change this linear production model (where we extract materials, elaborate products and dispose them), circular economy (CE) emerges as a restorative and regenerative model, which aims to keep products (as well as their components and materials) at their highest utility and value for as long as possible (Ellen Macarthur Foundation, 2015). Circular principles involve technical and biological cycles, where the latter consists of flows of renewable biotic resources, aiming to design products consumed or used under a cascading approach, and subsequently biodegraded to re-enter the biosphere and begin a new cycle (Bocken et al., 2016). CE in the context of the use of biomass for bio-based products, known as the circular bioeconomy, suggests first addressing

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biomass towards the production of materials before (after one or more cascading steps) its final management in energy recovery or composting processes (known as the waste hierarchy concept).

The manufacturing of apple fruit, such as juice production, is one example of the intensive waste generation during the industrial stage of this product. The main by-product generated corresponds to apple pomace, a heterogeneous mixture of peel, core, seed, calyx, stem, and soft tissue (Aghili et al., 2019; Adil et al., 2007). In fact, it is estimated that around four million tonnes of apple pomace are produced annually (Gołębiewska et al., 2022). However, the apple pomace has been so far sold for animal feeding or sent to landfills or incinerators. In the latter, it results in economic costs for disposal, leading to negative effects on the environment (such as greenhouse gas emissions and underground water pollution) and incurring into a loss of value with the wasting of large amounts of valuable nutrients (Gołębiewska et al., 2022), vitamins, dietary fibre and phenolic compounds (Skinner et al., 2018; Wang et al., 2019).

Despite its high moisture content of apple (70–85 %), its rich composition in lignocellulosic components, with values of 7–44 % cellulose, 4–24 % hemicellulose, and 15–23 % lignin (Costa et al., 2022), makes possible the manufacturing of products useful in other downstream processes of the bio-based value chain. Nevertheless, the creation of new valorisation pathways must be accompanied by a technological development. The design of the biorefineries must adapt to the feedstock to be transformed into high value-added products while keeping its independence on fossil fuels, increased productivity, and resource security (Rebolledo-Leiva et al., 2022a).

Although the large volume of waste generated from the apple manufacturing industry, there is limited research on the topic which implies a technological shortcoming for its implementation (de Oliveira et al., 2022). The currently studied apple pomace-based biorefineries were aimed at the manufacturing of products like acrylic acid (Okoro et al., 2022), n-butanol and electricity generation (Molina-Guerrero et al., 2022), and ethanol production (Glucose → Ethanol + CO<sub>2</sub>) (Borujeni et al., 2022). It is perhaps the last one the more versatile product, because of the countless industrial applications of ethanol as solvent, fuel as well as its use as intermediary in the production of other chemicals (Joseph et al., 2023). Even though the studies done on biorefineries for the valorisation of apple pomace have been framed mainly within the energy sector, other alternatives with a focus on the extraction of polyphenols may be proposed. This is because pomace retains phenolic compounds, which is explained by the low content (3–10 %) in apple juice (Ferrentino et al., 2018). Besides, the high potential for the recovery of polyphenols is also motivated by their increasing demand, due to their health benefits related to the prevention of cardiovascular diseases, diabetes (type II), cancers, and its antioxidant properties (Li et al., 2020).

The traditional procedure for the extraction of phenolics involves the use of organic solvents, for instance 70 % acetone or 80–100 % methanol solutions to extract them from apple pomace (Diñeiro García et al., 2009; Suárez et al., 2010). When these types of organic solvents are used, they must be removed from the product and recycled again to be reused in the process (Garrido Makinistian et al., 2019). Its flammable and toxic characteristics encourages the search and use of others which may be Generally Recognized as Safe (GRAS) by the European Food Safety Authority (EFSA) for food applications, like water and ethanol (EFSA, 2012; Castro-Puyana et al., 2017). As green solvent, the use of water is considered a satisfactory choice for phenolic compounds extraction due to its safety, accessibility, and low cost (Reis et al., 2012). Furthermore, the Soxhlet technique is a standardized method commonly used, e.g., in the food and pharmaceutical industries, for solid-liquid extraction (Chemat et al., 2012; Peanparkdee and Iwamoto, 2019), which uses a mixed and separate solvents (Rashd et al., 2024) for extracting analytes from solid samples. This method is favoured over other leaching techniques due to its superior effectiveness (Torres-Rodriguez et al., 2024). Its advantages are that it is simple and

inexpensive, but its main barrier lies on the requirement long processing times (e.g., up to 72 h) to reach high yields (Skendi et al., 2022). Nevertheless, there have been developments to reduce the extraction time and the amount of solvent consumption (Torres-Rodriguez et al., 2024).

The great attention paid in the literature to the extraction and recovery of phenolic compounds from agri-food waste is evidenced by multiple extraction techniques (i.e., maceration, decoction, percolation, infusion, digestion and serial exhaustive) with the laboratory scale being the mainstay for designing industrial-scale production routes (Alara et al., 2021). In the case of the apple pomace, the reviews by Zhang et al. (2021) and (Awasthi et al., 2021) identified different methods for extracting its phenolic compounds such as supercritical/subcritical CO<sub>2</sub> extraction, ultrasound-assisted, and solvent (acetone, methanol) extraction, with varying yields. However, the experimental processes should demonstrate the environmental performance before its implementation at industrial scale. Thus, this manuscript aims to propose a biorefinery design that maximizes the utilization of apple pomace for the co-production of value-added bioproducts, such as phenolic compounds and ethanol. For this, process modelling and life cycle assessment (LCA) are combined to identify the hotspots that may represent environmental barriers at an early stage of biorefinery design, as well as the phases that contribute most throughout the bioproduct life cycle. In addition, based on the systematic literature review performed (see Table S1 of Supplementary Materials), the motivation for this manuscript lies in the fact that no articles were identified about the LCA of valorisation pathways of apple pomace focusing on the extraction of phenolic compounds. The main products analysed were electricity, digested, compost, succinic acid, protein ingredients, fattening food and ligninolytic enzymes.

This research presents a first environmental insight for achieving an efficient use of apple pomace for the joint production of ethanol and extraction of total phenolic compounds. For the latter, due to the various extraction techniques tested in the literature, this manuscript compares two methods: “green” extraction with solvents (mainly water) and the Soxhlet technique (conventional method). In this regard, the question to be answered by the LCA is which design could be more attractive, since the Soxhlet method requires ethanol solvent that could also be produced by the biorefinery (reducing its demand), or whether it is preferable to sell both products to the market. The design of the platform will be assumed to be in Chile, as this country is the main exporter of apples in the southern hemisphere, and it ranks fourth worldwide (after China, USA, and Italy) accounting 9.5 % of world exports (Iriarte et al., 2021). Furthermore, apple pomace represents 13.5 % of the total apple production in Chile, equal to >164,000 tonnes of the harvest fruit (Hernández et al., 2021). Traditionally in this country, apple pomace is left on agricultural land or used for animal feeding. Although the feed strategy is relevant in the circular economy model, this research aims to propose a maximisation of the use of pomace biomass to obtain multi-products for different industries such as food or pharmaceuticals (with TPCs), and for energy or chemical (with bioethanol). In this way, this research report contributes with the environmental assessment of a multiproduct design of apple pomace-based biorefineries to promote a low carbon and circular food industry.

## 2. Materials and methods

### 2.1. Biorefinery modelling

The biorefinery design has an annual processing capacity of 4000 tonnes of apple pomace, equivalent to the pomace production of one of the main apple manufacturing plants in Chile, in which the potential biorefinery is assumed to be integrated. The platform consists of four main stages: pre-treatment, fermentation, purification for ethanol production, and phenolic compound extraction. In the last one, two configurations are considered for the biorefinery since the phenolic compounds are extracted through water (see Fig. 1) and Soxhlet (see

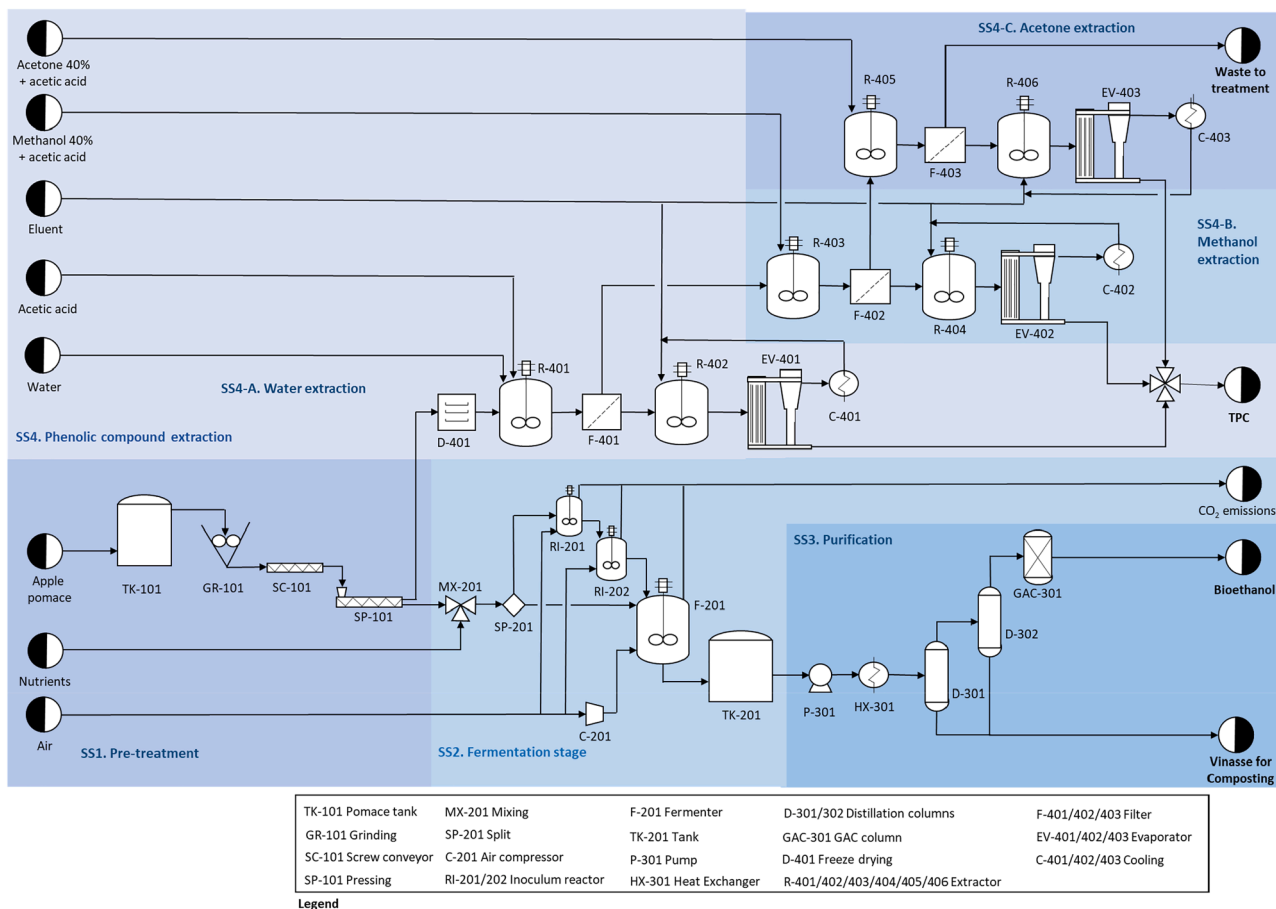


Fig. 1. Flow diagram of ethanol production and TPC extraction through solvents.

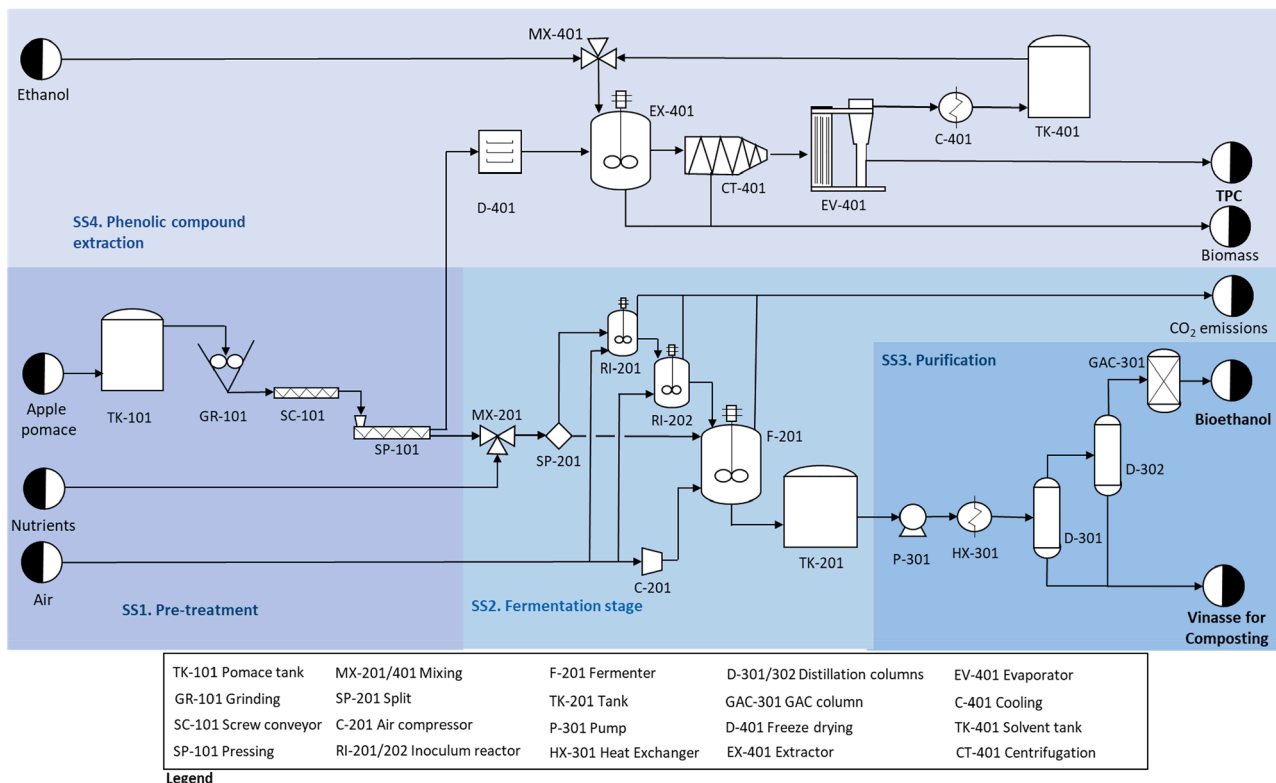


Fig. 2. Flow diagram of ethanol production and TPC extraction through Soxhlet method.

Fig. 2) extraction methods. The process was modelled in the Superpro designer® software v11 (Intelligen Inc., 2023).

### 2.1.1. Bioethanol production

The modelling of the second-generation (2G) bioethanol production from apple pomace was based on the work performed at lab-scale by Hernández et al. (2021). In the pre-treatment stage, the pomace was milled to obtain a homogenised flow and pressed to extract the free liquid phase, which subsequently passes through the fermentation section. The solid phase was afterwards fed to the facility section in charge of the phenolic compound's extraction. In the fermentation, it was assumed that 5 % of the fermentable sugars are used for inoculum preparation (i.e., yeast production). Nutrients were included at  $0.4 \text{ g} \cdot \text{L}^{-1}$  and the reaction was performed at  $30 \text{ }^\circ\text{C}$  during 144 h (Hernández et al., 2021). In the purification stage, distillation columns were used for ethanol recovering and purification. The upgraded ethanol was the distillate while the raffinate was a vinasse subproduct in need of further processing. After distillation, ethanol dehydration was performed to generate fuel-grade ethanol with a product purity of 99.5 %. On the other hand, the apple vinasse raffinate was treated following a composting strategy. For this, the inventory data was obtained from a previous work of our group carried out by Estévez et al. (2023).

### 2.1.2. Total phenolic compound extraction

Two strategies have been analysed to extract the total phenolic compound from the solid flow obtained from the pressing unit: i) solvents extraction and ii) Soxhlet extraction. The first is selected because water at room temperature is the main agent used for the extraction, which represents a "green" technique. The second is selected because it is a well-known technique with good efficiency, as mentioned above.

**2.1.2.1. TPC extraction with solvents.** The extraction of phenolic compounds was modelled following the fractionation method performed by Reis et al. (2012). Accordingly, the apple pomace is freeze drying and stirred with water at room temperature during 90 min. Then, the water extracts are filtered, and the residual pomace is reconstituted in methanol solution (40 %) and stirred as above. The same procedure was lastly repeated with acetone (40 %) on the residue left from the methanol extraction. Besides this, acidic conditions were used in both cases (addition of acetic acid at  $5 \text{ mL} \cdot \text{L}^{-1}$ ) to avoid oxidation of phenolic compounds. Phenolic-rich fractions were eluted with methanol containing 0.1 % HCl and were concentrated using an evaporator at  $40 \text{ }^\circ\text{C}$  until achieve a powder with a purity of 95 %. According to Reis et al. (2012), the fractionation of 1 kg of dried apple pomace yield extracts equivalent to about 2.566 g of gallic acid. The water extraction procedure reaches 67 % efficiency in total phenolic compounds (TPC), followed by the extraction of 17 % and 16 % of TPC using methanol and acetone, respectively.

**2.1.2.2. Soxhlet extraction.** Ethanol was used for Soxhlet extraction of TPC from freeze dried apple pomace (with a moisture content of about 19 %) using the technique described by Ferrentino et al. (2018) and Paes et al. (2014). An advantage of the Soxhlet extraction is that a filtration process to recover the solvent containing extracted phenolics is not needed (Al Jitan et al., 2018). This method consisted of 150 mL of solvent over 5 g of apple pomace sample in a Soxhlet equipment during 6 h at the boiling temperature of the solvent. The use of ethanol as solvent is well-known for its great affinity for the extraction of antioxidant (Ferrentino et al., 2018). In the process modelling, a 5 % of ethanol losses through air emissions were assumed following the work of Barjoveanu et al. (2020). The obtained extract from Soxhlet method was centrifuged and then the supernatant was put in a evaporator (Ferrentino et al., 2018). Soxhlet technique achieves a yield (i.e., the mass of the extract and the mass of the sample used for the extraction) of 47.4 % (w/w dry basis) and about 4.13 mg of gallic acid equivalent per g

of extract (Ferrentino et al., 2018). The waste biomass generated is assumed to be treated for composting.

## 2.2. Life cycle analysis

The life cycle analysis was performed under the ISO 14,040–14,044 guidelines (ISO, 2006a, 2006b), which is a four-step standardised methodology: aim and scope of the study, life cycle inventory, impact assessment, and interpretation.

### 2.2.1. Aim and scope

As mentioned above, this work aims to compare two biorefinery designs that maximizes the use of apple pomace for the production ethanol and the extraction of phenolic compounds. For this purpose, the LCA methodology is useful to support the decision-making process for a possible implementation of the design. The study followed a *cradle-to-gate* attributional approach (see Fig. 3), considering all the activities corresponding to the raw material extraction (i.e., background processes of energy, chemicals, and other consumables), apple cultivation and manufacturing, and bioproducts elaboration. The functional units (FU) used for reporting the environmental loads were expressed in terms of mass (1 kg) in the case of bioethanol, and 1 kg of equivalent gallic acid extracted for TPC, as they are commonly used in the literature. A volume unit (L) for bioethanol was also considered for the discussion of results in Section 3.3.1. In the case of total phenolic content, these are usually determined by the Folin-Ciocalteu method, and the results are expressed in gallic acid equivalents (Carlqvist et al., 2022), which also motivated the selected FU. These FUs are selected for comparative purposes with previous studies in the literature. For example, LCA studies such as those of Frascari et al. (2019), Carlqvist et al. (2022), and Salzano de Luna et al. (2023) addressing the extraction of phenolic compounds used mass-based FUs.

### 2.2.2. Life cycle inventory (LCI)

The foreground LCI was built following a "bottom-up" approach and thus considering the outcomes from the mass and energy balances of the apple pomace biorefinery design, using the Superpro designer® software v11 (Intelligen Inc., 2023). The results are presented in Table 1 for the bioethanol production, Tables 2 and 3 for phenolic compounds with water and Soxhlet extraction, respectively. The electricity production of Chile was modelled considering the energy profile of the year 2023 (CNE, 2024). Furthermore, the supply of steam was assumed to come from cogeneration systems to avoid the consumption of fossil resources. The background processes were taken from the Ecoinvent® v3.9 database, considering also the apple cultivation stage in Chile (Wernet et al., 2016). The transport distance of fresh apple to the gate of the biorefinery was 300 km, in accordance with the literature (Hajjaji et al., 2013; Lijó et al., 2017).

Regarding the manufacturing stage of apple, the LCI of juice concentrate production was taken from the work of Cheng et al. (2022). Orchards and the apple manufacturing plant are in Maule region in the Central Valley of Chile (specifically in the location of Romeral). Thus, fresh apples transport to the manufacturing plant and apple pomace to the biorefinery were neglected, as it was considered that the biorefinery will be coupled to the apple juice manufacturing plant. In addition, the distribution of the loads between the main product and the apple pomace in the manufacturing stage was done with an economic allocation. An average market price of  $1.5 \text{ } \$ \cdot \text{kg}^{-1}$  for the apple juice during the period 2018–2022 (ODEPA, 2023) and  $0.01 \text{ } \$ \cdot \text{kg}^{-1}$  for the apple pomace according to the information provided by the manufacturing plant. Thus, allocation factors are 99.5 % for apple juice and 0.5 % to apple pomace. Furthermore, although the vinasse treatment section produces a compost that could be considered as product from the composting process, all the burdens were allocated to the main two products (i.e., bioethanol and TPC). In addition, in the pre-treatment section, a mass allocation was used to distribute the loads of the

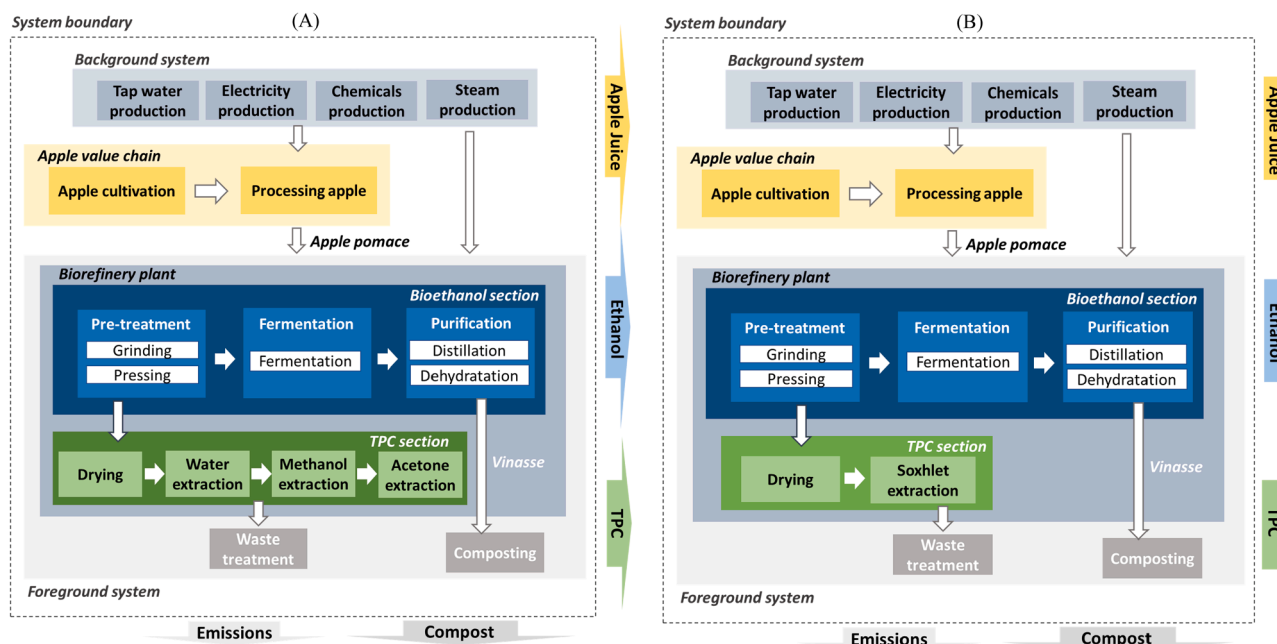


Fig. 3. System boundary of the 2G bioethanol and phenolic extraction with solvents (A) and Soxhlet technique (B).

Table 1

Life cycle inventory of ethanol production (FU: 1 kg).

Section of the facility	Inputs from Technosphere	Value	Outputs to Technosphere	Value
SS1. Pre-treatment	Pomace (kg)	14.26		
	Electricity (MWh)	0.29		
SS2. Fermentation	Nutrient (kg)	0.32	CO <sub>2</sub> emissions (kg)	1.15
	Water (kg)	18,35		
	Chilled water (kg)	222,63		
	Electricity (MWh)	1.73		
SS3. Purification	Cooling water (t)	1.31	Ethanol (kg)	1.0
	Steam (kg)	13.18	Vinasse (kg)	12.20
	Chilled water (kg)	8.29		
	Electricity (MWh)	0.003		

pomace, as it is an intermediate biomass flow without economic value. The allocation corresponds to 25 % for TPC and 75 % for ethanol. Finally, the translation of LCI data into environmental impacts was carried out using the SimaPro® v9.4.0.2 software (PRé Consultants, 2020).

### 2.2.3. Life cycle impact assessment

To transform the inventory data to environmental impacts, the LCIA characterisation factors from the ReCiPe 2016 (H) V1.07 / World (2010) (H) method (Huijbregts et al., 2017) were considered. The selection of the method lies in providing a mid-point impact analysis, considering characterisation factors that can be representative for a Chilean context. As there are no South American factors yet, only global methods such as IMPACT World or ReCiPe are available, the latter being one of the most widely used due to its frequent updating (Borghesi et al., 2022). Hence, the impact categories selected were Global Warming (GW), Stratospheric Ozone Depletion (SOD), Particulate Matter (PM), Terrestrial Acidification (TA), Freshwater Eutrophication (FE), Marine Eutrophication (ME), Terrestrial Ecotoxicity (TET), Freshwater Ecotoxicity (FET), Human Carcinogenic Toxicity (HT); Land Use (LU) and Fossil Resource Scarcity (FRS). Following a systematic literature review (see Tables S4 and S5 in the Supplementary Materials), global warming, acidification, eutrophication and human toxicity were the most frequently used categories. On the other hand, given the agricultural origin of the biomass (i.e., from apple cultivation) to be valorised,

Table 2

Life cycle inventory of TPC extraction with solvents (FU: 1 kg of TPC).

Section of the facility	Inputs from Technosphere	Value	Outputs to Technosphere	Value
SS1. Pre-treatment	Pomace (kg)	764,62		
	Electricity (MWh)	15.14		
SS4-A. Water extraction	Water (t)	6.47	Wastewater (m <sup>3</sup> )	4.36
	Acetic acid (kg)	36.5		
	Methanol (kg)	103.7		
	HCl (kg)	1.05		
	Chilled water (t)	25.0		
	Steam (kg)	608.0		
SS4-B. Methanol extraction	Electricity (kWh)	147.8		
	Methanol (t)	2.69	Wastewater (m <sup>3</sup> )	5.87
	Water (t)	3.88		
	Acetic acid (kg)	48.6		
	HCl (kg)	1.02		
	Chilled water (t)	115.5		
	Steam (t)	1.15		
	Electricity (kWh)	1.52		
SS4-C. Acetone extraction	Acetone (t)	2.59	TPC (kg)	1.00
	Water (t)	3.88	Residual biomass (kg)	873.0
	Acetic acid (kg)	54.9	Wastewater (m <sup>3</sup> )	6.64
	Methanol (kg)	103.7		
	HCl (kg)	1.05		
	Chilled water (t)	25.0		
	Steam (kg)	249.4		
	Electricity (kWh)	1.35		

Table 3

Life cycle inventory of TPC extraction with Soxhlet technique (FU: 1 kg of TPC).

Section of the facility	Input from Technosphere	Value	Outputs to Technosphere and Nature	Value
SS1. Pre-treatment	Pomace (kg)	318,23		
	Electricity (kWh)	6.3		
SS2. Soxhlet extraction	Ethanol (kg)	962.6	TPC (kg)	1
	Steam (t)	2.6	Residual biomass (kg)	916,45
	Cooling water (t)	238.86	Ethanol (air emission) (kg)	331.91
	Electricity (kWh)	105.32		

resource scarcity and land used were selected. Similarly, in the case of phenolic compound extraction, solvents and energy may be relevant for the ozone depletion category. Finally, a sensitivity analysis was carried out after the estimation of the environmental burdens and the identification of most impactful product system stages.

### 3. Results and discussion

#### 3.1. Environmental profiles

The overall outcomes per scenario proposed for the LCA for the 2G bioethanol production and TPC extraction from apple pomace are displayed in Table 4. Besides these results, a relative contribution analysis per process stage was provided in Fig. 4 aiming at the identification of the most concerning ones for the environmental profile. As shown for the bioethanol production (Fig. 4a), all the stages could be potential hotspots since their relevancy differs from one impact category to the other. The first section to be highlighted is fermentation which stands out (32 %–48 %) in five of the eleven selected indicators (PM, FE, FET, HT, and FRS). The reason behind this is the electricity demand in the reactor, which is caused by the large residence time of the process (144 h).

In addition, the composting treatment of vinasse is the second most important stage, as it is the hotspot in categories such as GW (77 %), SOD (98 %), and TA (67 %). Direct emissions are the main responsible items of the burdens in the above mentioned three categories. Finally, purification and pre-treatment were only relevant in two (TET and LU) and one (ME) category respectively. In the purification, distillation is the unit most impactful because of the use of steam, while the consumption of apple pomace is the main hotspot in the pre-treatment stage. This is due to the upstream processes of apple production, such as cultivation.

In the case of the TPC extraction with solvents (see Fig. 4b), the subsequent use of organic solvents to increase the amount of phenolic extracted is the critical factor in the environmental performance. Although the proportion of acetone used (40 %) is lower than traditional doses (e.g., 70–80 %), the acetone extraction is the most impactful stage (between 49 %–87 % for ME and HT, respectively) in almost all categories evaluated. The exception occurs in SOD (with 56 %) and LU (with 52 %) categories, where methanol extraction is the hotspot process.

**Table 4**  
Environmental profile of bioethanol and TPC production from apple pomace.

Impact category	Unit	Bioethanol (FU: 1 kg)	TPC extraction with solvents (FU: 1 kg)	TPC with Soxhlet (FU: 1 kg)
Global Warming	kg CO <sub>2</sub> eq	3.17	5887	219
Stratospheric Ozone Depletion	g CFC <sub>11</sub> eq	0.06	1.27	0.61
Particulate Matter	kg PM <sub>2.5</sub> eq	0.01	6.89	1.55
Terrestrial Acidification	kg SO <sub>2</sub> eq	0.02	18.58	2.04
Freshwater Eutrophication	kg P eq	0.001	1.50	0.16
Marine Eutrophication	kg N eq	0.0001	0.15	0.014
Terrestrial Ecotoxicity	kg 1,4-DCB	3.73	7379	1868
Freshwater Ecotoxicity	kg 1,4-DCB	0.02	56.20	5.94
Human Carcinogenic Toxicity	kg 1,4-DCB	0.04	92.46	14.01
Land Use	m <sup>2</sup> a crop eq	1.62	11.20	538
Fossil Resource Scarcity	kg oil eq	0.27	4023	60.71

Regarding to the acetone extraction, the profile is primarily defined by the consumption of acetone with 10 categories of 11 (see Fig. 5a). It is only in LU where the demand of acetone is no longer the main environmental concern but rather the use of process steam in the evaporation unit. This is related to the burning of wood resources in the cogeneration. Similarly, to the extraction with acetone, the environmental profile with methanol is also characterised by the use of the solvent. As shown in Fig. 5b, the exception is LU, on which the use of steam is the outstanding contributor.

In comparison with the previous technique, Soxhlet extraction implies a significant reduction of loads with a decrease that may reach values as high as 96 % (GW) and 98 % (FRS). Other remarkable improvements were observed in eutrophication categories with 90 % both. In contrast with these favourable outcomes and in line with those of the preceding method, the LU category has reported results influenced by the steam demand with a rise of impact of about 79 %.

In the contribution analysis of Soxhlet alternative (see Fig. 4c), two are the main concerning factors. On the one hand, steam demand highlights in most of categories such as GW, SOD, TA, TET, and LU categories with values from 53 % to 98 %. On the other hand, electricity stands out in the PM category (65 %), while both factors (i.e., electricity and steam) in categories such as HT and FRS have similar contributions. Finally, the ethanol emissions are relevant in the human toxicity category, representing about 42 % of the impacts.

#### 3.2. Sensitivity analysis

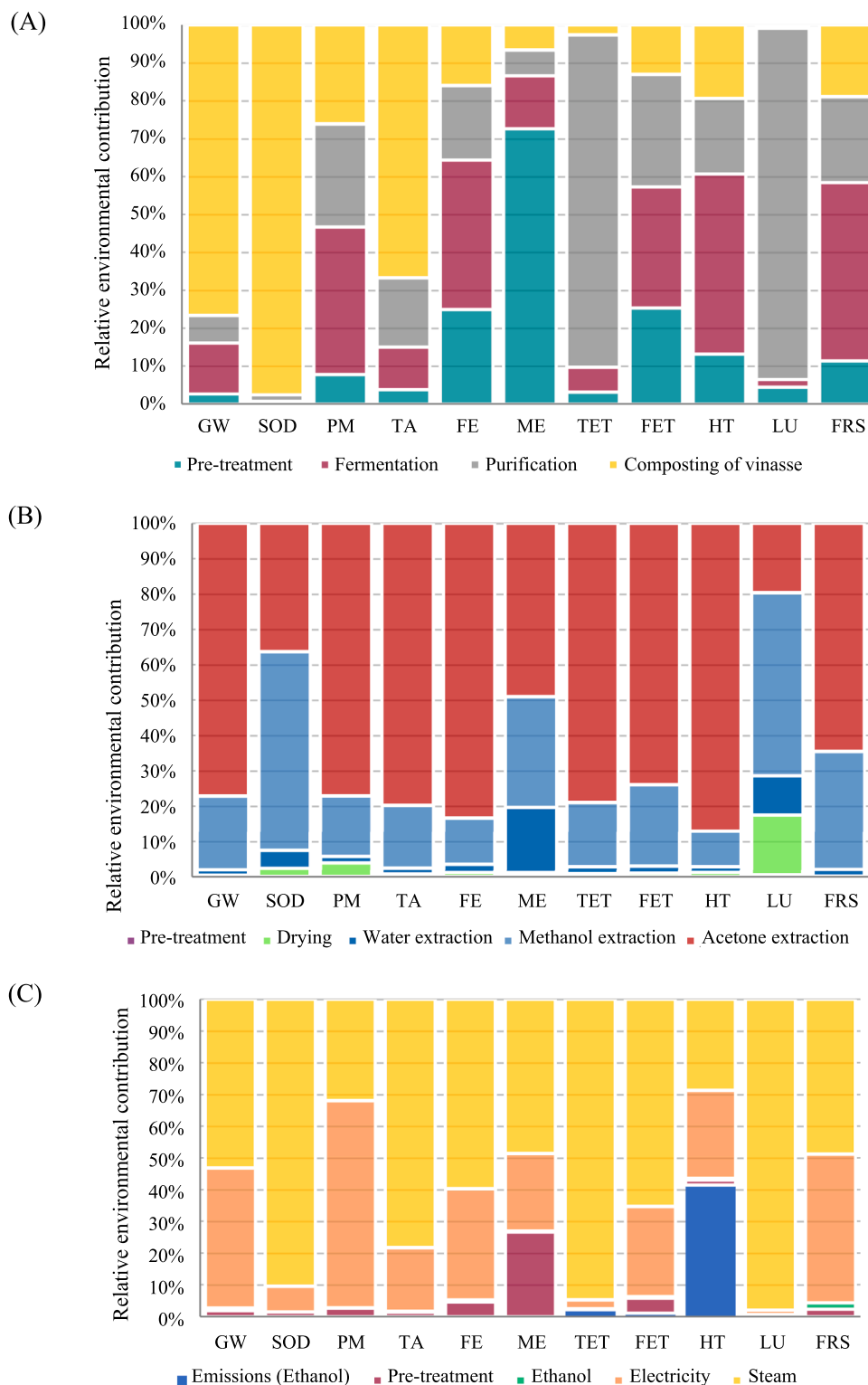
Based on the identification of the critical stages that may limit the potential implementation of this apple pomace-based biorefinery, a sensitivity analysis is performed considering the avoidance of the use of organic solvents (methanol and acetone) in TPC extraction, changes on the allocation factor of the apple pomace, and on the electricity mix of Chile.

##### 3.2.1. TPC extraction through solvents

According to Reis et al. (2012), phenolic compounds of apple pomace (mainly phenolic acids and flavonoids) are readily extracted with water, but the subsequent use of organic solvents is expected to rise the efficiency of the process. According to these authors, the extraction yield with methanol and acetone after water extraction is 0.44 and 0.41 g of gallic acid per kg of dried apple pomace, respectively. However, both values are rather low compared to the standalone extraction with water (0.17 g of gallic acid per kg of dried apple pomace), which does not compensate the higher environmental impacts of organic solvents. Thus, to improve the environmental performance of the TPC extraction, an alternative could be the use of water and methanol as extractants. The goal is to estimate the level of importance of the induced changes on the environmental performance of TPC extraction. In relationship with the results achieved and shown in Fig. 6, the GW indicator is expected to reduce up to 73 % using the water–methanol extraction procedures, reaching a value of 1600 kg CO<sub>2</sub>eq·kg<sup>-1</sup> of TPC. Furthermore, avoiding only the use of acetone has a low reduction in SOD (24 %) and LU (5 %) indicators regarding the baseline, due to the relevance of methanol in these burdens. On the other hand, if the use of both organic solvents (i.e., methanol and acetone) was avoided, the GW could be 178 kg CO<sub>2</sub>eq·kg<sup>-1</sup> of TPC, which is equivalent a reduction of 97 % respect to the baseline (i.e., with three extractions procedure: water, methanol, and acetone). Other relevant improvements are achieved in TA and HT categories, where the lack of use of organic solvents decreases the impacts by 96 %, and 97 % in FRS. Thus, the largest decrease in most of the categories analysed is reaching using only water extraction.

##### 3.2.2. Allocation factor of apple pomace

The selection of the allocation method is relevant for reporting the environmental profile of a bio-based product or when the aim is to compare it with other alternatives, e.g., fossil-counterpart



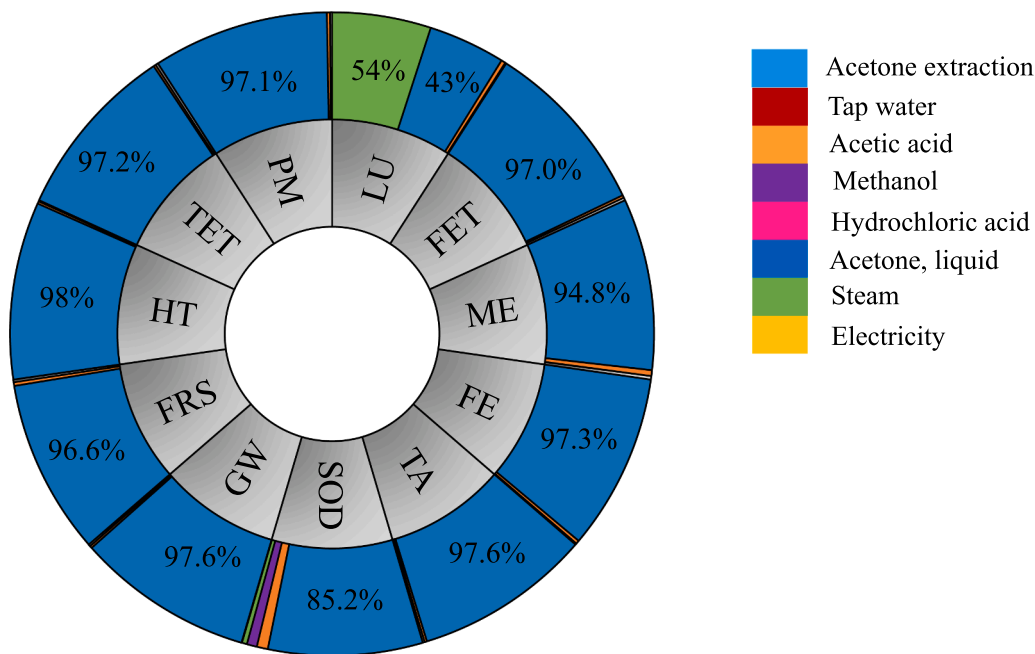
**Fig. 4.** Relative contribution analysis of bioethanol production (A) and TPC with solvents (B) and Soxhlet (C) extraction. Emissions in (C) correspond to the loss of ethanol in the form of air emissions.

(Rebolledo-Leiva et al., 2022b). However, the use of the economic allocation can be influenced by the volatile market prices of products or feedstocks. Therefore, to provide an overview about how the market growth induces changes in the allocation factors, potential variations of 10 % and 20 % were assumed. These market fluctuations have been measured with the use of the producer price index (PPI) and are in line with those that has occurred in Europe, which has ranged between 8 %

and 15 % in the period 2021–2022 (European Central Bank, 2023). The PPI has been used instead of the consumer price index (CPI) since it is able to tack the manufacturing prices of the industry, before importations and taxes, and without considering the living costs of the population.

In the case of bioethanol production (see Fig. 7a), an allocation factor of 20 % for the apple pomace may increase its GW profile about 14 %

(A)



(B)

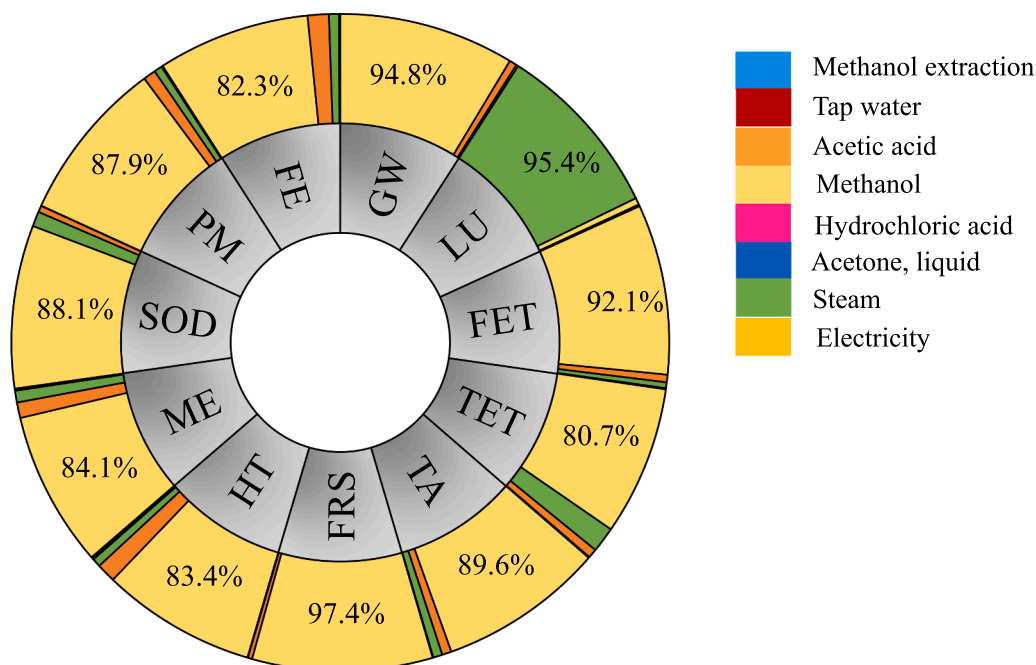


Fig. 5. Contribution analysis in the acetone (A) and methanol (B) in the solvent extraction process.

(equivalent to 3.70 kg CO<sub>2</sub> eq). Despite being the global warming, the recurrently addressed environmental indicator in the assessment of products and processes, the ME category is the most affected impact category. A rise of 96 % with respect to the baseline scenario, reaching a value of 3.05 g N eq per kg of ethanol, has been estimated. FET and FE also growth significantly with a proportion of 89 % and 88 %, respectively. The lowest increase was observed in the SOD indicator with about 9 %, as the greatest contributor is the treatment or valorisation of the vinasses with composting.

Regarding the TPC extraction using organic solvents (Fig. 7b), the influence of the background emissions of the pomace production are not

significant. In line with this, only two categories (ME with 15 % and LU with 17 %) were the most affected. In the case of Soxhlet extraction (Fig. 7c), the consumption of pomace is also significant in ME with a notably increase of about 91 % with respect to baseline. On the other hand, indicators such as FET (59 %) and FE (57 %) are also influenced by apple pomace reaching greater impacts due to the increased allocation factor.

### 3.2.3. Electricity mix generation of Chile

In 2022, renewable sources reached a 55.6 % share in Chile's electricity generation (CEN, 2023). To unveil how the use of a lower

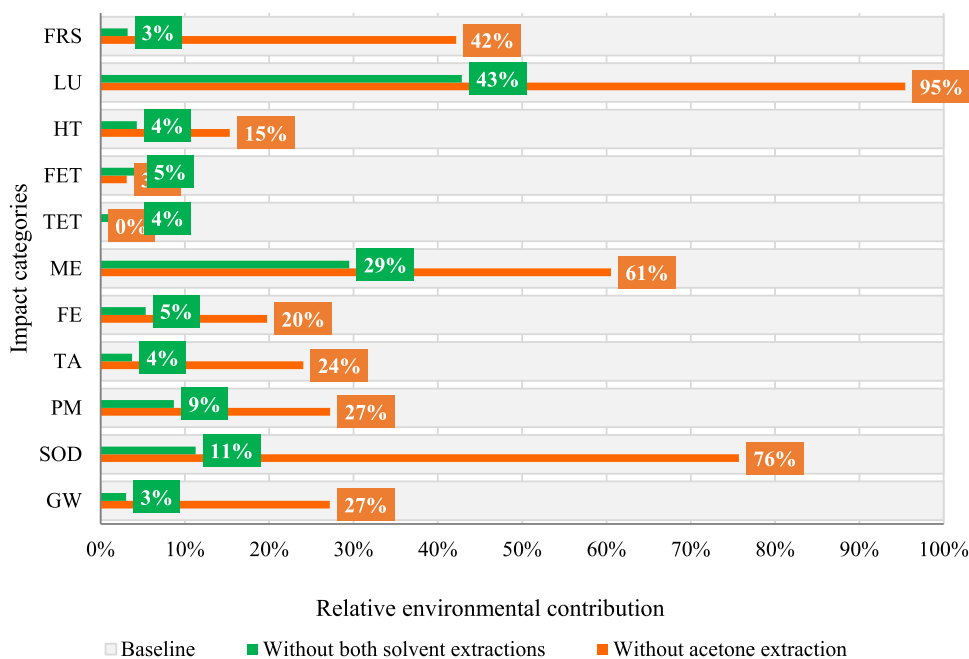


Fig. 6. Sensitivity analysis of TPC profile without organic solvent extraction.

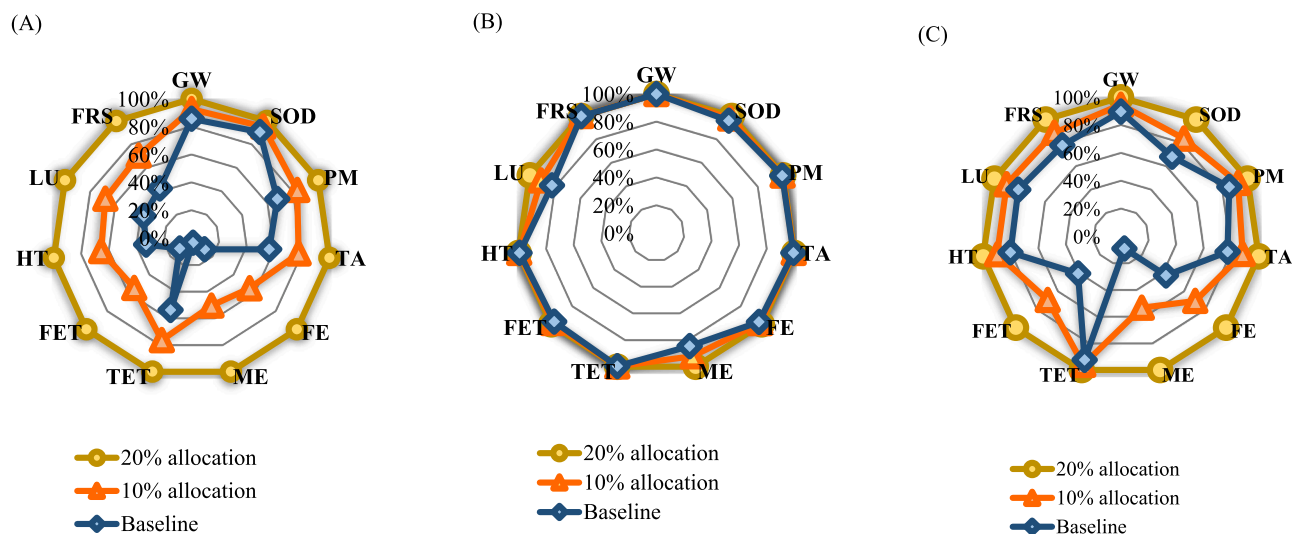


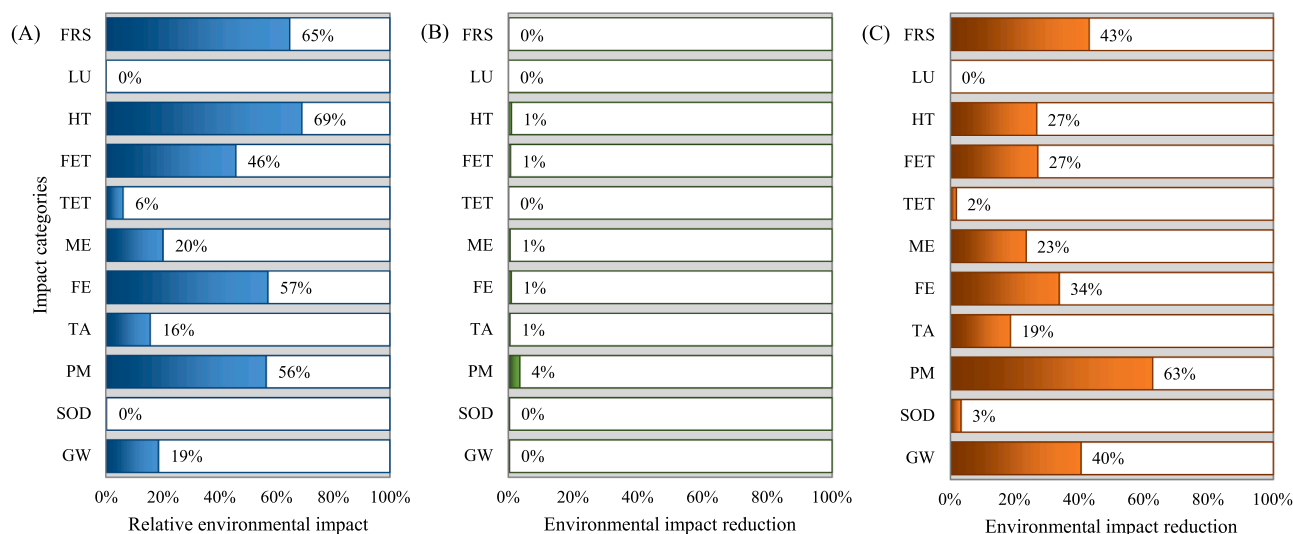
Fig. 7. Variation of allocation factor of apple pomace on the environmental profile of ethanol (a) TPC with solvents (b) and Soxhlet (c) extraction.

environmentally electricity profile influences the results, the design of the manufacturing process for ethanol and the extraction of polyphenols have also contemplated the consumption of a full-renewable electricity (energy target by 2050). This electricity mix has been assumed to be characterised by 40.7 % solar, 30.5 % hydro, 25.4 % wind, 0.5 % geothermal, and 2.9 % biomass energy. This hypothetical scenario has been selected based on the historic data of the electricity generation of Chile (CEN, 2023). During the period 2017–2022, the largest increase in contribution has centred on solar (from 5 % to 17 %) and wind (from 5 % to 11 %), while hydro has ranged between 20 %–30 %, biomass around 2 %, and geothermal around 0.1–0.5 % share.

Under these considerations, the results indicate that the bioethanol profile (see Fig. 8a) is reduced significantly in almost all categories. For instance, the most affected are HT (69 %), FRS (65 %), and FE (57 %). On the other hand, the widely well-known GW encompasses a value of 2.59 kg CO<sub>2</sub> eq per kg of product, which is equivalent to a reduction of 19 %. The exceptions are the indicators of LU and SOD with very slight

variations (0.1 %–0.2 %).

Concerning phenolic compounds, the alternative of the extraction with solvents (see Fig. 8b) is not affected by the change of the electricity mix, as the main contributors are the background emissions from the production of the organic solvents. The category presenting the largest reduction is PM although with only 4 % with respect to the baseline scenario. On the other hand, the extraction of TPC with the Soxhlet technique (see Fig. 8c) is improved using renewable-based electricity. For instance, the GW profile is about 131 kg CO<sub>2</sub> eq per kg of TPC extracted, which is equal to a reduction of 40 % regarding the baseline scenario. The largest decrease levels are observed in categories such as PM (63 %) and FRS (43 %), while minor variations are identified in LU (0.1 %), TET (2 %), and SOD (3 %).



**Fig. 8.** Effect of renewable electricity on the environmental profile of ethanol (a), TPC with solvents (b) and Soxhlet (c) extraction. The colour bars indicate the environmental reduction from the baseline (which is 100 % in A, B and C) and the scenario using renewable energy.

### 3.3. Comparison with the literature

#### 3.3.1. Ethanol production

In the literature, the production process of bioethanol has been widely studied for a large variety of feedstock and valorisation routes. Examples could be the research performed by [Munoz et al. \(2014\)](#), [González-García et al. \(2018\)](#), [Lyu et al. \(2020\)](#) and [Wang et al. \(2013\)](#). In the respective order of publications, the following feedstocks were evaluated: maize grain, maize stover, sugarcane, sugar beet and wheat; barley straw and brewer's spent grains; cassava root, cassava straw and whole plant cassava; and sweet potato. Different outcomes and conclusions have been reached for the climate change category. While [Munoz et al. \(2014\)](#) reported values in the range of 0.7 to 1.5 kg per kg ethanol under a *cradle-to-gate* approach, which are lower than the 3.17 kg CO<sub>2</sub> eq obtained here, however, no treatment of vinasse subproduct is addressed in that study. Higher GW values are also reported in the literature such as the work performed by [González-García et al. \(2018\)](#), who reached a value of 7.39 kg CO<sub>2</sub> eq per kg product with similar system boundary. According to these authors, autohydrolysis treatment and production of enzymes caused the highest impacts.

Regarding to the outcomes of [Lyu et al. \(2020\)](#), a GW profile of 1.74, 2.93, 1.55 kg CO<sub>2</sub> eq per L of ethanol was estimated using as feedstock cassava root, cassava straw, and the whole plant cassava, respectively. In this study, the waste liquid is assumed to be sent to an anaerobic fermentation and subsequently to a wastewater treatment unit. [Wang et al. \(2013\)](#) determined a value per L of product of 1.47 kg CO<sub>2</sub> eq based on sweet potato, and steam consumption during bioethanol conversion was the critical factor, as also occurs in this manuscript. Based on density of 789 kg·m<sup>-3</sup>, the GW profile in this manuscript is 2.5 kg CO<sub>2</sub> eq per L of bioethanol, which may be higher in some cases, but is within the range of these literature studies.

#### 3.3.2. Phenolic compounds extraction

When Soxhlet extraction is considered, the production of total phenolic compounds from the apple pomace involves 219 kg CO<sub>2</sub> eq per kg of TPC. Other studies that also approaching the Soxhlet method with different feedstocks is the work of [Arias et al. \(2022\)](#), which found that about 90 and 200 kg CO<sub>2</sub> eq per kg of antioxidant-rich powder (95 % purity) were emitted in the extraction of phenolics from leaves and stem of beet wastes, respectively. Similarly, [Santiago et al. \(2021\)](#) considered the Soxhlet method for extracting the main bioactive compound (i.e., rutin) from asparagus residues. Per kg of rutin, a value of about 1450 kg of CO<sub>2</sub> eq under an economic allocation (between the rutin product and

biofertiliser) or 146 kg CO<sub>2</sub> eq with a mass allocation.

Other studies used different extraction methods such as [Carlqvist et al. \(2022\)](#), who calculated a GW profile of 0.68 kg CO<sub>2</sub> eq per kg of phenolic compounds extracted from spruce bark, using a hot water extraction procedure. Similarly, techniques such as supercritical fluid extraction (SFE), pressurised liquid extraction (PLE), maceration and ultrasound assisted extraction (UAE), and water extraction have proven to have similar environmental profile. For example, the first three methods were ranged by [Arias et al. \(2022\)](#) in about 60 to 900 kg CO<sub>2</sub> per kg of powder product. [Carlqvist et al. \(2022\)](#) found out that UAE and SFE reached 11 and 5.8 kg CO<sub>2</sub> eq·kg<sup>-1</sup> total phenolics, respectively. The reason for these results is mainly the environmental burden caused by the consumption of ethanol in UAE and SFE processes. This also demonstrates the relevancy of the proper selection of solvents for phenolic extraction. Finally, higher GW impacts were reported with SFE method by [Santiago et al. \(2021\)](#) (2039 kg of CO<sub>2</sub> eq per kg of rutin) and [Rodríguez-Meizoso et al. \(2012\)](#) (2364 kg of CO<sub>2</sub> eq per kg of rutin), although it is relevant to consider that rutin is only one of the total phenolic compounds.

### 3.4. Future perspectives

The biorefinery design proposed contributes to support the transition towards a sustainable production pattern (i.e., the Sustainable Development Goal 12). This is motivated as the aim is to valorise a by-product from the food industry, and subsequently, maximise the use of the residual biomass.

The ethanol produced from the liquid stream after the pressing unit in the biorefinery could be used as solvent for the TPC extraction route to reduce its demand. However, the quantity produced in this design is very low compared to the ethanol demanded by the Soxhlet technique. The amount of ethanol produced is equivalent to about 13 % of the quantity demanded by the TPC extraction. Therefore, further research could take two approaches. On the first hand, a higher ethanol production yield could be reached with another feedstock that present a higher proportion of fermentable sugars. Secondly, the mixture of the on-site ethanol with other coming from another biorefinery platform. Although the consumption of this solvent is not a critical parameter in the environmental profile of the production of phenolic compounds, the design of an integrated biorefinery, under cascade approach, could be attractive for optimising the demand of resources. Potential challenges of these strategies could be logistic issues such as supply planification, location of suppliers, and transport. Thus, to overcome them decision-

makers could be supported with optimisation tools to identify feasible solutions considering environmental, economic and social goals.

Since the composting stage is the greatest impact in bioethanol production, the search for other treatment strategies for vinasse should be the focus of attention. Without considering the composting, the GW profile of bioethanol is about 1.07 kg CO<sub>2</sub> eq per kg of product (70 % lower). Thus, the selection of the vinasse treatment is significantly relevant for ensuring a low environmental performance of the biorefinery platform. In addition, it could be interesting to analyse the feasibility of using the residual biomass obtained from the extraction of TPC for animal feed as another co-product of the system.

Since many biorefineries are still in the development phase, primary data from scaled processes is scarce or commercially sensitive to use in LCA studies (Gaffey et al., 2024). In this regard, it is important to mention that a level of uncertainty exists in projecting scale-up calculations. In this research, average data were used from the experimental procedure collected from the literature. Thus, future research could focus on this issue to determine the variations in the environmental profile of the modelled bioproduct on an industrial scale. As a complement to life cycle analysis, future research of the co-production systems of ethanol and polyphenolic compounds could also include economic aspects for the determination of the feasible scale of production to reach the minimum selling price that can be competitive in the market for both products. Finally, the environmental effects of using apple pomace for value-added bioproducts instead of its traditional use as animal feed can be addressed with a consequential LCA approach in future studies.

#### 4. Conclusions

This research presents the combination of process modelling and the environmental analysis for the co-production of bioethanol and total phenolic compound extraction for an apple pomace-based biorefinery. The aim was to identify the steps that contribute most to environmental performance in the design phase to promote new bio-based circular models that reduce dependence on fossil resources. The main findings suggest that the composting treatment of the vinasse is the critical contributor to the environmental profile of bioethanol. In the extraction of phenolic compounds, the use of organic solvents, such as methanol, and especially acetone, are the most impactful factors of this route. This advocates the use of the Soxhlet technique, which significantly reduces the environmental loads of TPC extraction, achieving a more competitive GW profile representing a difference of about 96 % compared to the use of solvents. In the Soxhlet route, demand of electricity and steam were the most impactful factors. Thus, the moving towards a renewable based electricity generation can notably decrease the environmental profile of TPC through this pathway. Although the environmental profiles of both products are within the range values of the related literature, further studies could estimate the economic viability and the verification of an alternative vinasse treatment to improve the environmental and economic attractiveness of this multiproduct biorefinery design.

#### CRedit authorship contribution statement

**Ricardo Rebolledo-Leiva:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Sofia Estévez:** Writing – review & editing, Visualization, Methodology. **Diógenes Hernández:** Writing – review & editing, Resources, Investigation, Formal analysis. **Gumersindo Feijoo:** Writing – review & editing, Validation, Supervision. **María Teresa Moreira:** Writing – review & editing, Validation, Supervision. **Sara González-García:** Writing – review & editing, Validation, Supervision, Project administration, Funding acquisition.

#### 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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#### Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.clcb.2024.100125](https://doi.org/10.1016/j.clcb.2024.100125).

#### Data availability

Data will be made available on request.

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