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Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace

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ABSTRACT

To determine the environmental and economic performance of emerging processes for the valorization of red wine pomace, a techno-economic assessment (TEA) and a Life Cycle Assessment (LCA) are combined at an early design stage. A case study of two polyphenol extraction methods at laboratory scale, solvent extraction (SE) and pressurized liquid extraction (PLE), were first analyzed via a carbon footprint (CFP). Subsequently, the laboratory scale design was improved and translated into industrial scale and a TEA was performed on the industrial scale designs. Finally, LCA was applied again with all impact indicators and the information gathered from both the TEA and LCA was combined into concise decision support, using Multiple Criteria Decision Analysis (MCDA). SE performs better than PLE, due to a lower solvent to DW ratio and a less expensive processing setup in both environmental and economic terms. The CFP of at laboratory scale aided in showing potential environmental hotspots and highlighted the need to reduce solvent use. The MCDA showed a shift in decision support depending on how strongly economic or environmental benefits are valued and eases the interpretation of the 19 different indicators derived from the TEA-LCA results. Both SE and PLE with a solvent to dry weight (DW) ratio of 5 and 10, respectively, perform competitively while SE with a solvent to DW ratio of 10 outperforms PLE with a solvent to DW ratio of 25. The case study illustrated how early design calculations (CFP), and combined LCA and TEA may be combined to improve process design.

1. Introduction

Biomass demand for the production of bioenergy, biomaterials and biochemicals is estimated to increase by 70–110% by 2050 compared to 2005 levels (Mauser et al., 2015). A paradigm shift to renewable sources of production has long been discussed, in the context of circular economy and valorization of biomass waste resources produced through the agricultural value chain. The bioeconomy today is estimated to have a €2.4 billion annual turnover, and it is only expected to increase in the future (Scarlat et al., 2015). Yet, the prefix bio does not guarantee sustainability. For example, growing biomass for biofuels has long been debated (Haberl et al., 2010; Murphy et al., 2011; Popp et al., 2014), prompting the Renewable Energy Directive (The European Commission, 2018) at an international, pan-European, level to ensure valid quantification of greenhouse gas reductions claims. In this regard, integration of methods such as life cycle assessment (LCA) and techno-economic assessment (TEA) are valuable input for quantitative sustainability assessments.

Combined TEA-LCA has been applied in many occasions to assess the environmental and economic ramifications of implementing new technologies (Cai et al., 2018; Hise et al., 2016; Vaskan et al., 2018). More interestingly, TEA-LCA has been used to quantify and monetize externalities, namely environmental damages, to provide a more complete picture of the financial burdens arising from environmental problems (Ögmundarson et al., 2018; Pizzol et al., 2015). Recently, combined TEA and LCA has been used to optimize new production routes from an early design phase, as in the case of integrated wastewater treatment and microalgae production for biodiesel production (Barlow et al., 2016), or the integration of power-to-gas technology of methane and photovoltaics (Collet et al., 2017). Combined TEA and LCA lends itself well to

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finding production hot spots and opportunities for optimization. This is even more relevant when applied to renewable resources such as biomass, which have to be managed sustainably.

New materials like biodegradable bio-sourced biopolymers and bioactive molecules such as, polyphenols obtained from agricultural residues can be combined to create new and innovative products (Vannini et al., 2019). Polyphenols present interesting possibilities as they can be utilized by various industries, such as in the pharmaceutical, nutraceutical and cosmetic industries (Pérez-López et al., 2014). Among other, polyphenols have been shown to have excellent health promoting qualities, such as anti-diabetic, anti-inflammatory, anti-bacterial and anti-cancer properties (Nowshehri et al., 2015). This versatility means that polyphenols may be used in niche markets as well as in mass markets, with various uses that may be of importance to the bioeconomy e.g. active packaging, coloring, food supplements, etc. Wine pomace is a residue rich in polyphenols, with a global production of 68 million tons of wine pomace annually (Nowshehri et al., 2015). To ensure a sustainable exploitation of polyphenol rich biomass, innovative polyphenol extraction methods at the laboratory scale were analyzed using TEA-LCA in order to identify hotspots and potentially environmentally problematic production steps.

On the other hand, results from the application of TEA-LCA can sometimes be confounding if, for example, one option performs better environmentally while incurring financial loss. The multitude of factors that must be taken into account remains an issue, when policy makers, corporations, or any other actor is faced with the need to decisively and definitively choose between alternative solutions to a given problem. In order to handle this, the decision-making context surrounding such a choice can be handled in many ways, from community-based decision making to round table discussions or even executive fiat. But, without a tool for interpreting fundamentally conflicting information, the results of the decision making process can vary wildly and may depend on happenstance and or subjective factors. Multiple Criteria Decision Analysis (MCDA) has been applied to aid in alleviating these problems by introducing a transparent and repeatable form of decision support (Kalbar and Das, 2020; Köksalan et al., 2011).

When assessing environmental issues in an LCA perspective, oftentimes practitioners turn to single indicators such as global warming potential (carbon foot-printing), but this poses potential downfalls such as burden shifting e.g. shifting environmental burdens from carbon emissions to environmental or human toxicity (Laurent et al., 2012). In other cases, practitioners turn to endpoint damage modeling, but these have high levels of uncertainty, can lead to unintentional bias (Kalbar et al., 2012a; Sohn et al., 2017), and still leave the decision maker with several categories of environmental damages e.g. ecosystem health, human heath, and resource availability. Furthermore, neither of these methods can be directly combined with economic indicators. In some cases, LCA practitioners have monetized impacts in order to combine environmental and economic indicators, however these suffer from issues, among others, involving the relationship of internalized and externalized costs (Reap et al., 2008). These issues have lead some LCA practitioners to turn to MCDA for providing decision support (Kalbar et al., 2016, 2012a; Sohn et al., 2017), as applying MCDA with a defined decision context to results from TEA-LCA is advantageous when a final decision must be taken.

Therefore, in this study LCA is applied at an early design stage to obtain a preliminary carbon footprint (CFP) of the polyphenol extraction methods. Subsequently, the design of the laboratory extraction procedures is improved and adapted to industrial scale and a TEA of the industrial scale scenarios is performed. Then LCA is applied again with all environmental indicators in simulated industrial conditions. This is done with the goal of obtaining a holistic picture of the economic feasibility and possible environmental impacts of each polyphenol extraction method. Lastly, MCDA is applied to the decision context of choosing between the polyphenol extraction methods and a weighting-profile derivation method (ArgCW-LCA) is applied (Sohn et al., 2020). The criteria from the LCA and TEA are incorporated to provide concise decision support for selecting one of the laboratory methods for scale-up.

2. Material and methods

Results of laboratory scale experiments of different methods for the extraction of polyphenols from red wine pomace were evaluated using a combination of TEA and LCA. Two different labs, one located at the University of Bologna, Italy, and a second located at the Research Institute of Sweden (RISE), provided operational parameters for their laboratory setups. Yields, solvent amounts, temperature and time were then used to complete the inventory to carry out a preliminary carbon foot-printing (CFP) LCA of the laboratory scale experiments. The parameters of the most successful setups i.e. those producing the highest polyphenol yields, were used for the CFP and are described in detail in Table S1 of the supplementary information. The laboratory methods are described briefly in Section 2.1. Following this step, industrial scale processes of the laboratory methods were designed and optimized for key parameters, using TEA (described in Section 2.3). An LCA of the optimized industrial scale processes including all environmental indicators was then carried out. Lastly, a multiple criteria framework for decision support where the economic and environmental indicators are combined was applied to the results from the TEA-LCA.

2.1. Polyphenol extraction methods and laboratory experiments

The CFP of two different extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE), was determined. One SE setup and 3 different PLE setups, where the main difference is the solvent amounts used, were assessed for this step, of which the most successful setups in terms of yield are briefly described below, and the remainder can be found in the SI, since they did not become relevant for the industrial case. The laboratory extraction methods are also described in detail in (Ferri et al., 2020).

2.1.1. Solvent extraction with acetone

Solvent batch extraction was performed in the laboratory with various solvents (acetone, ethanol, and aqueous aceto-nitrile), temperatures (50 or 70 °C), and extraction times (1, 2 or 4 h). Of all operational parameters tested, an SE with the following conditions attained the highest polyphenol yield (Ferri et al., 2020). Solvent extraction with 61% acetone, and 39% water as solvent on a per mass basis, with a solvent to dry weight (DW) ratio of 11:1. Extraction was performed in an air-tight vessel at 50 °C at atmospheric pressure where the solvent and previously ground pomace were kept in contact for 2 h. Due to the polarity of polyphenols, they easily solubilize in polar media such as water/organic solvent and hydro-alcoholic mixtures. Once solubilized, polyphenols are carefully extracted from the liquid phase using a rotary evaporator under vacuum conditions, since many phenols also exhibit thermal instability. A powder is obtained from the rotary evaporator, which is then analyzed for polyphenol content of the extracts. Polyphenol content is expressed in kg gallic acid equivalents (kg GAE).

2.1.2. Pressurized liquid extraction with ethanol

As with SE, various operational conditions were tested for PLE. An ethanol/water (EtOH—H2O) mixture was used in combination with CO_2. The ratios of EtOH—H_2O:CO_2 varied from 75% to 50% and 100% in the various conditions tested, while the contact time tested varied from 30, 40 and 50 min (Ferri et al., 2020). PLE performed with 37% ethanol, 39% water and 25% supercritical CO_2 on a per mass basis was shown to attain the highest yield between the operational conditions tested. The extraction was performed at 80 °C and 100 bar, at this temperature and pressure CO_2 is in the supercritical region, according to its phase diagram. As this is a continuous set up, where the solvent flows through the vessel containing the pomace, it leads to a high solvent to

DW ratio of 101.

2.2. Carbon foot-printing of laboratory scale experiments

A CFP was performed on one SE and 3 PLE extraction methods, using only the Global Warming potential (GWP) impact category as the environmental indicator. The ReCiPe 2016 Midpoint Hierarchist (H) method (Huijbregts et al., 2017), which has a 100 year time horizon from point of emission, was used as impact assessment method, supplied by the Ecoinvent 3.4 Database (Wernet et al., 2016) and processed with the open source software OpenLCA (GreenDelta, 2019). The functional unit for the CFP is the production of 1 kg of polyphenols in kg GAE, assuming equal functionality. The process design software, SuperPro Designer v.10 (Intelligen Inc, 2018), was used to simulate the polyphenol extraction methods with industrial scale equipment. The operational parameters that attained the highest polyphenol yields in the laboratory (SE with acetone with a DW of 11:1, at 50 °C, for 2 hrs and PLE with 75% EtOH:H2O, 25% supercritical CO_2 at 80 °C and 100 bar) were used for the CFP, as well as 2 other PLE shown only in the SI, Table S1. These operational parameters were used to populate the SuperPro Designer model so as to obtain the rest of the inventory of for example, energy and heat consumption, needed for the CFP. For the most part, the lab set up was kept the same. Through consultation with project partners it was possible to identify industrial scale equipment that would be able to perform the same functions as equipment in the lab, e.g. a spray dryer with nitrogen instead of a rotary evaporator for isolation of the polyphenols, distillation equipment for solvent recovery, etc.

The polyphenol producing plant is assumed to be placed in Italy and thereby, background processes for Italy from the Ecoinvent database were used as much as possible, e.g. the electricity grid.

2.3. Conceptual design of industrial scale processes

The process design focused on optimizing the operational parameters of the laboratory extraction methods so that it would be economically feasible to implement a polyphenol extraction at industrial scale. In order to achieve this, solvent recovery and product concentration are essential i.e. several process steps are required such as distillation, nano filtration, and spray drying (Fig. 1 and Fig. 2). The solvent loss and the energy required for solvent recovery should be reduced as much as possible. The solvent to DW ratio is an important parameter in solvent recovery. Industrial scale extraction processes usually have multiple extraction stages in a counter current flow setup (Berk, 2018). This setup reduces the required solvent to DW ratio and increases the product concentration in the extract, which reduces both the solvent recovery costs and the product concentration costs.

Based on literature (J.A. Dávila et al., 2017a; J.A. 2017b; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017), process setups were designed for both SE (Fig. 1) and PLE (Fig. 2). Both designs assume multiple extraction stages in counter current flow. Compared to the laboratory scale experiments the residence times were adjusted as well as, flow and equipment sizes. The total extraction time is assumed to be 60 min for all processes. As shown in Ferri et al. (2020), the effect of lengthening extraction time was low on total polyphenol yields, thereby the yields obtained for 1 hour or 2 hrs of extraction are comparable. This is why it was deemed possible to obtain the same polyphenol yields for SE even with a 60 min residence time. Likewise, the authors found that a doubling of the acetone content for SE did not attain enough improvement of the polyphenol yield to justify the extra solvent use at industrial scale (Ferri et al., 2020).

A set up with counter current flow allows for a reduction of the solvent to DW ratio used in the laboratory scale experiments, while the extraction yield, i.e. the amount of polyphenols extracted per kg DW, is maintained. As mentioned previously the solvent to DW ratio is an important parameter. The reduction of the solvent to DW ratio in the industrial scale processes is difficult to estimate precisely, therefore, based on J.A. Dávila et al. (2017a; J.A. 2017b); Fiori (2010); Todd and Baroutian (2017); Viganó et al. (2017) and expert knowledge from the collaborating laboratories (Ferri et al., 2020), three feasible solvent to DW ratios were used in the TEA and LCA for each extraction method. The parameters of these scenarios are shown in Table 1. In all scenarios, the amount of polyphenols extracted is assumed to be equal to the laboratory scale experiments, since total residence times and solvent amounts are mostly within the ranges tested in the laboratory, though for a few of the scenarios it is important to validate the yields by further experiments i.e. SE-2 and, PLE-10 and PLE-25, which are assumed to attain the high yield due to the countercurrent set-up (Berk, 2018). The solvent to DW ratios and the solvent compositions were corrected for the amount of water in the pomace. The number in each scenario name refers to the solvent to DW ratio.

The designs of both extraction processes include grinding of pomace to increase contact with the solvent, multiple extraction stages, distillation for solvent separation and recovery, nano filtration to concentrate the polyphenols, and finally spray drying for recovery of the polyphenols in powder form (Fig. 1 and Fig. 2). The solvent to DW ratio determines the concentration of polyphenols after extraction and

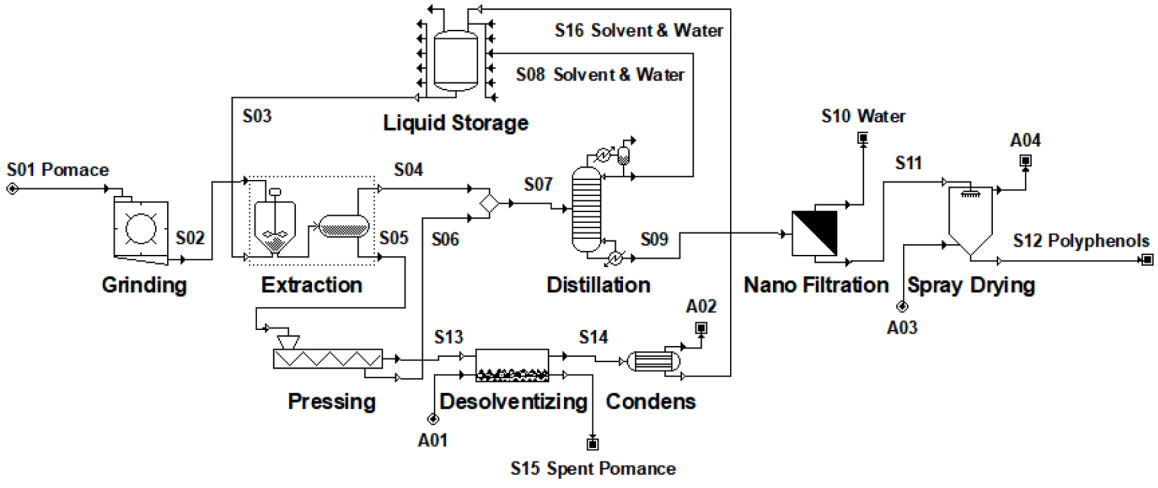

Fig. 1. Process flow diagram for solvent extraction with acetone and water, for polyphenol recovery from grape pomace. Process includes input of wine pomace (S01), grinding, addition of solvents (S03) from liquid storage, extraction of polyphenols, distillation for solvent recovery and recycle (S08), nano filtration and spray drying for concentration and final recovery of polyphenols (S12), pressing and desolventizing of the wet pomace, condensation for additional recovery of solvent from the soaked pomace (S16).

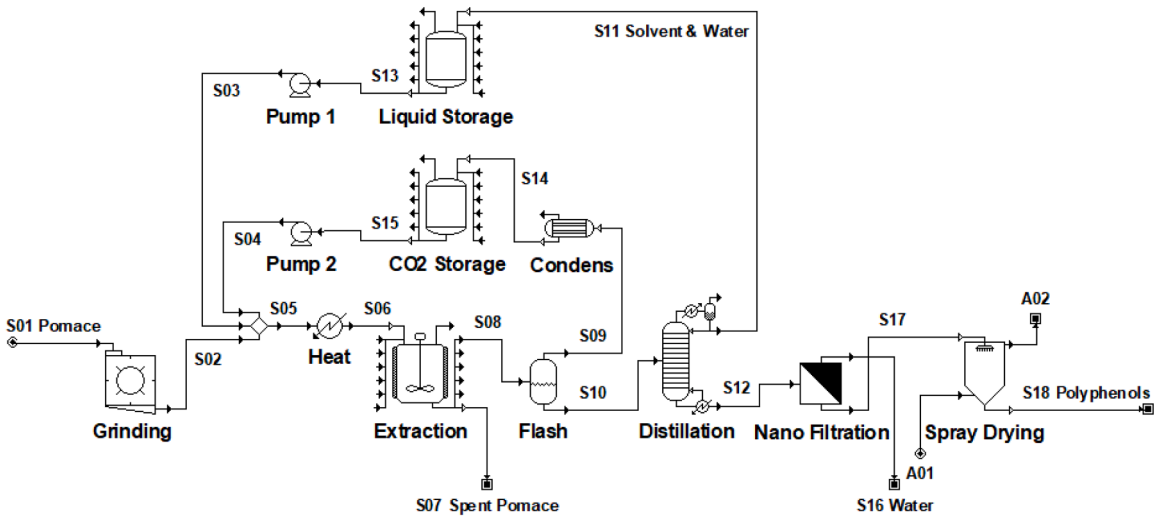

Fig. 2. Process flow diagram for pressurized liquid extraction with ethanol, water, and supercritical CO_2 for the extraction of polyphenols from grape pomace. Process includes input of wine pomace (S01), grinding, pressurization by pump 1 and 2, addition of liquid solvents (S13) from liquid storage and supercritical CO_2 (S15) from CO_2 storage, extraction of polyphenols, flashing for CO2 recovery (S09) and distillation for liquid solvent recovery and recycle (S11), nano filtration and spray drying for concentration and final recovery of polyphenols (S18). Spent pomace (S07) is not desolventized.

Table 1
Design parameters for industrial scale processes used in TEA and LCA.

	SE-10	SE-5	SE-2	PLE-50	PLE-25	PLE-10
Solvent to DW ratio (kg/kg DW)	10	5	2	50	25	10
Extraction stages	2	2	5		2	
Residence time (min/stage)	30	30	12		30	
Polyphenols extracted (g GAE/kg DW)		47			79	
Temperature (°C)		50			80	
Pressure (bar)		1			100	
Composition solvent						
- Water		33.3%			37.5%	
- Acetone		66.7%			–	
- Ethanol		–			37.5%	
- CO_2		–			25.0%	

distillation i.e. the higher the solvent use the lower the polyphenol concentration in the liquid. The extracted polyphenols after distillation are concentrated i.e. water is removed, by nano filtration to 25% DW and then to 95% DW by spray drying.

For SE, the solvent is recovered from the pomace by first pressing i.e. separating the majority of the solvent from the pomace and distilling the liquid fraction, while the pomace is sent to desolventizing (drying). The composition of the solvent in the recycle is 95% acetone and 5% water. For scenario SE-2, it is necessary to dry the pomace prior to extraction, because otherwise the required solvent composition cannot be obtained. This dryer is not shown in Fig. 1, but is taken into account in the TEA and LCA.

For PLE, the solvent is recovered from the pomace by flashing the CO_2 and distilling the extract. The composition of the solvent in the recycle is assumed to be 90% ethanol and 10% water.

2.4. Techno-economic assessment of industrial scale processes

TEA of the designed industrial scale processes was carried out in order to investigate the economic repercussions of installing a polyphenol extracting plant. The TEA includes Capital Expenditure (CapEx) and Operating Expenditure (OpEx). Assumptions and simplifications were made in order to fill in data gaps. The most important assumptions considering the TEA are reported in Table 2. Assumptions of economic parameters were based on Intelligen Inc, (2018); Maroulis and Saravacos (2007); Peters et al. (2003); and Sinnott and Towler (2009).

Based on the flow sizes of the designed processes, equipment were scaled. Purchased equipment cost and CapEx were based on the literature used for the process designs (J.A. Dávila et al., 2017b; J.A. 2017a; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017) and the references mentioned above. The CapEx of the extraction vessels was scaled using the six-tenths factor (Maroulis and Saravacos, 2007; Peters et al., 2003; and Sinnott and Towler, 2009). and was corrected for pressure (see detailed estimations in Table S2 of the SI).

In several wine growing areas wine pomaces and other residues are currently processed on industrial scale in centralized processing plants, so called distilleries. It is assumed that the polyphenol extraction will be performed in a setting similar to that of existing distilleries e.g. as in Italy and France, where 100% and 90% of wine pomace is sent to distilleries for treatment, respectively (Galanakis, J.A. 2017). The raw material costs for the polyphenol extraction are assumed to be negligible, since pomace is already part the current residue processing system.

The labor related costs were assumed to be the same for all scenarios and were based on: 2 shift positions, 4.8 operators per shift position, and an operator salary of k€ 30/y. Costs for supervision, direct salary overhead, and general plant overhead are added to the costs for operating labor.

Maintenance, including tax, insurance, rent, plant overhead, environmental charges, and royalties are assumed to be 10% of the CapEx per year. The financing costs are based on an amortization of the CapEx

Table 2
Parameters for the techno-economic assessment.

Production hours	8000	h/y
Red wine pomace	20	kton wet/y
- fresh wine pomace	2500	kg wet/h
- dry weight percent	36%	DW
Polyphenols extracted		
- with SE	340	ton GAE/y
- with PLE	572	ton GAE/y
Labor related costs	891	k€/y
Maintenance, etc.	10%	of CapEx/y
Financing costs	10%	of CapEx/y

over 10 years with no interest (Peters et al., 2003; Sinnott and Towler, 2009).

The heat and electricity required in the different processes was based on process simulations in SuperPro Designer and on process parameters described in Maroulis and Saravacos (2007); Peters et al. (2003); and Sinnott and Towler (2009). Utility consumption was generalized to facilitate the techno-economic evaluation, thereby the heat required in the dryer for SE-2 and in the spray dryer, as well as the energy required for solvent recycle is assumed to be two times the heat of evaporation of the concerning stream, based on process simulations with the flow sizes of the designed industrial scale processes. For SE, this energy is distributed as follows: 90% for distillation (heat) and 10% for desolventizing (heat). For PLE, this energy is distributed as follows: 90% for distillation (heat), 5% for pumping (electricity), and 5% for heating prior to extraction. The electricity usage of the processing units is assumed to be: 10 kWh/ton input for grinding, 5 kWh/ton input for pressing, 5 kWh/ton permeate for nano filtration, 10 kWh/ton input for spray drying (atomization). Cooling water is used for cooling, for which the costs are assumed to be negligible. Despite all measures in the designed processes to recover the solvent, solvent loss is inevitable. Therefore, for all scenarios, a solvent loss of 2% of the solvent in the recycle is assumed. Prices, CO_2-equivalents, and heat of evaporation of relevant utilities and solvents are given in Table 3.

2.5. Life cycle assessment of industrial scale processes

Following the TEA, an accounting LCA was performed on the newly designed industrial systems as modelled by the TEA. The functional unit is the production of 1 kg of polyphenols expressed as 1 kg GAE. The assessment is a "gate-to-gate" LCA and includes all actions carried out in order to obtain polyphenols from red wine pomace. This includes all steps from when the pomace enters the production system to the product, the polyphenols, leaving the production facility, e.g. all processing steps, such as grinding, drying, adding solvents, filtering, distillation and more (Fig. 1 and Fig. 2). The assessment does not include the end of life of the polyphenols or any transport throughout the life cycle, since this is deemed equal for all processing methods. Also, any potential burden of the raw material, the red wine pomace, is not accounted for, since the wine pomace is waste from wine production. Likewise, no credits are assigned for the production of polyphenols potentially replacing similar products in the market. The LCA includes all 18 impact categories in ReCiPe 2016 Midpoint (H) methodology (Huijbregts et al., 2017). The geographical location of the polyphenol plant is Italy.

2.6. Development of weighting for multi-criteria decision analysis

In order to incorporate the various environmental, as well as the economic criteria derived from the results from the previously described TEA and LCA assessments (see Section 3.2 and 3.3), the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) method of MCDA (Hwang and Yoon, 1981) is used. This method applies compensatory aggregation based on the definition of a positive ideal solution and a negative ideal solution, a theoretical best and worst case scenario respectively, and selecting the alternative with the shortest geometric distance from the positive ideal solution and the longest geometric distance from the negative ideal solution after weighting is applied for each criterion. This method of MCDA is chosen due to its previous application in the context of LCA and because it is one of the most widely applied compensatory methods of MCDA when cardinal indicators are available for all alternatives (Kalbar et al., 2012b; Kalbar and Das, 2020).

Table 3
Parameters for utilities and solvents.

		Price €/kWh	GWP CO_2-eq/kWh
Electricity		0.10	0.43
Heat		0.04	0.37
Cooling		0.00	0.56
Parameter	ΔH vap	Price	GWP
Unit	kJ/kg	€/kg	CO_2-eq/kg
Water	2260	0.00	0.0002
Ethanol	841	0.80	1.34
Acetone	539	1.20	2.87
CO_2	380	0.50	0.85

All midpoint indicators from LCA and production prices of the various polyphenol production methods from the TEA (Table S3) are used as criteria in the application of TOPSIS.

When applying TOPSIS, there is an inherent application of weighting, even in its default mode, equal weights are applied (Pizzol et al., 2017). This presents a problem because the selection of the ideal alternative is directly related to weighting, which is further discussed in section 4.1.1. In this case, following the ArgCW-LCA method (Sohn et al., 2020), normalization factors (NF) (PRé, 2019) per impact category (i) are used to derive a relative importance factor (RIF), relating the average value, amongst all of the alternative extraction methods, of each of the midpoint impacts (MI) to the average European's annual environmental impact such that $RIF_i = \overline{MI_i} \,/\, NF_i$ represents the relationship between environmental and other criteria (Equation 1). For example, for calculating the RIF_{GW}, if the average GW impact amongst all assessed technologies ($\overline{MI_{GW}}$), were 80 kg CO_2 eq., then because the NF_{GW} for GW is 7990 kg CO_2 eq., the RIF_{GW} will be approximately equal to 0.01. In this case, production cost is then normalized such that production cost is allocated the desired weight and the sum of all weights is equal to 1000. The resultant weighting is then displayed in tabular form to promote full transparency in the assessment (Table S4, and Table S5).

3. Results and discussion

3.1. Carbon foot-printing of laboratory scale experiments

The CFP analysis clearly shows that if laboratory conditions are maintained at industrial scale, then the acetone based solvent extraction method outperforms all other scenarios by a large margin, in terms of global warming potential (GWP), Fig. 3. This is largely due to the amounts of solvent used in each scenario, which are lowest for the Lab-SE-11 scenario. The large amount of solvent used in the continuous setup for all Lab-PLE scenarios results in a very high heating demand in, for example, heating during polyphenol extraction, and heating during distillation to recover the solvents.

From the CFP, the importance of keeping the solvent ratio as low as possible is evident. This has a trickledown effect on the energy demand of the whole system. The results can be used in the early design phase, in order to avoid excessive environmental burden later on. By identifying hot spots early on, it is possible to envision adjustments to the production setup, so that the identified hot spots are addressed. Measures, such as increasing the time of contact between solvent and pomace were identified after the CFP. Systems with multiple extraction stages and lower solvent to DW ratios were considered in the TEA.

3.2. Techno-economic assessment of industrial scale processes

The estimated CapEx for the different scenarios are: M€ 6.3 for SE-10, M€ 4.6 for SE-5, M€ 4.5 for SE-2, M€ 25.9 for PLE-50, M€ 16.6 for PLE-25, and M€ 9.8 for PLE-10. For the assessed solvent to DW ratios, the estimated CapEx are significantly higher for PLE compared to SE. Higher solvent ratios require larger equipment and a higher pressure results in more expensive equipment. Due to higher required solvent to DW ratios, the costs related to solvent recovery (i.e. electricity and heat) and solvent supplement are also higher for PLE compared to SE. On the other hand, PLE has a higher extraction yield compared to SE. By looking at

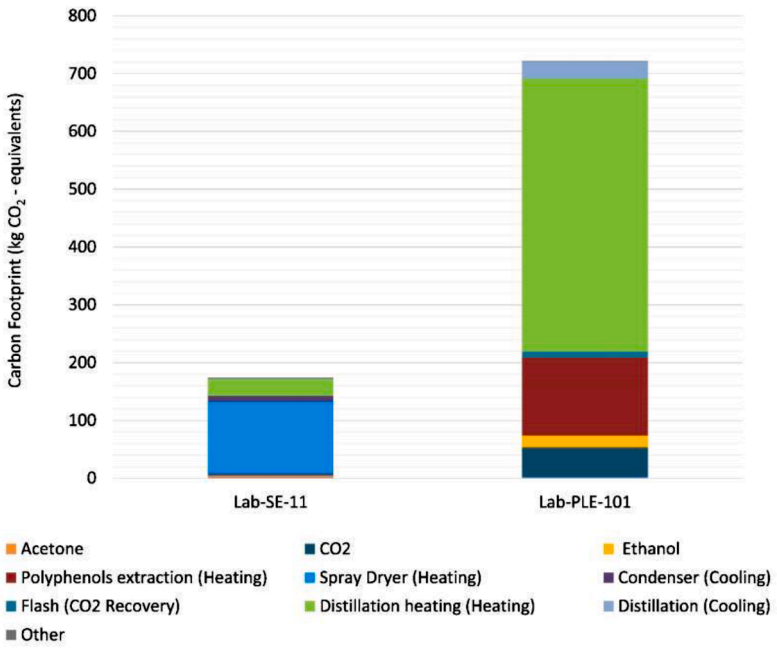

Fig. 3. Global warming potential results per kg GAE of polyphenol extraction scenarios at laboratory scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

processing costs expressed in €/kg GAE (Fig. 4), it is clear that the higher extraction yield for PLE does not compensate the higher costs. Only labor related costs are lower for PLE. Scenario SE-2, which has the advantage of a low solvent to DW ratio, has the lowest processing costs. However, because of the required drying step and the low solvent to DW ratio, the assumed extraction yield was considered to be uncertain. As a result, the most feasible options, from a techno-economic perspective, are SE-5 and PLE-10. In the technically feasible range of solvent ratios, SE performs techno-economically better compared to PLE. Details on estimated CapEx, solvent loss, and utility usage for all assessed scenarios is shown in Table S2 of the SI.

3.3. *Life cycle assessment of industrial scale design*

The LCA of optimized operational conditions showed that if seeking to alleviate GWP it would be preferable to choose SE-2, that is to say, a

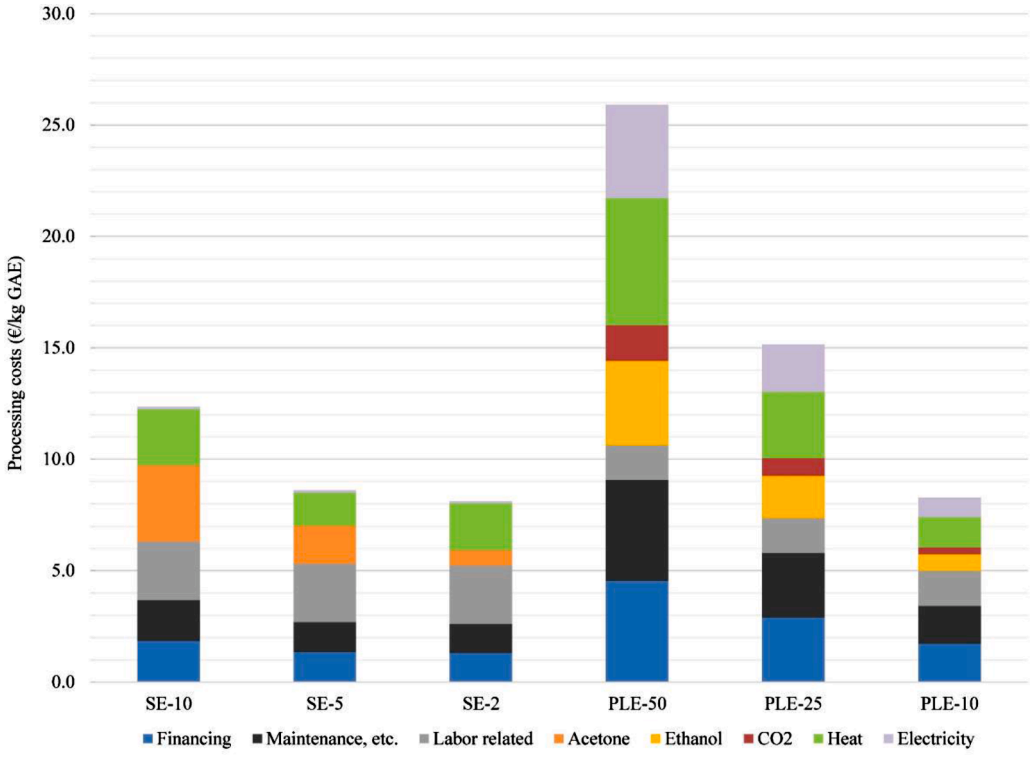

Fig. 4. TEA results of polyphenol extraction at industrial scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

solvent extraction using acetone with a solvent ratio of 2, Fig. 5. However, as mentioned previously, the extraction yield of SE-2 was considered to be uncertain and therefore SE-2 was not considered to be a competitive option. Moreover, PLE-50 and PLE-25 perform far worse than the other options in terms of GWP and all other impact categories (Figure S2, SI), so these are also not deemed competitive options.

From Fig. 5 it is possible to see the effect of the optimization performed via process design. The hotspot analysis still points towards solvent quantities as a key parameter for environmental outcomes, e.g. energy used for cooling and heating for distillation dominate the CO_2 burden, and energy for compressing the system. However, through process optimization it is possible to drastically reduce some GWP impacts that were large in the laboratory scale CFP, as for example the impact from the spray dryer for the SE options, by adding a concentration (filtration) step before the drying, which was not part of the laboratory design. On the other hand, it is possible to see that adding a drying step for the pomace in option SE-2, does not pay off in comparison to not drying in SE-5, as the dryer plus distillation heating and cooling, are on the same range of GW impact as just distillation heating and cooling in SE-5. The overall GWP is lower for all options due to the reduction in solvent use and addition of extraction steps.

Results of the TEA show the importance of the solvent to DW ratio for the feasibility of extraction processes. High use of solvent leads to high operational costs and increased demand for electricity and heat, which affect the results of both TEA and LCA. On the other hand, higher yields allow more leeway for higher energy consumption, though not always fully compensating for all GW impacts. A lower solvent to DW ratio results in lower costs for solvent recovery, lower solvent loss, and lower CapEx. These results are mirrored in the LCA, where results benefit from lower solvent use, while midpoint impacts are increased due to the extra heating demand from large solvent volumes. In this regard though, it was clear in the LCA that solvent use, especially if the solvent is acetone, comes with higher GW impacts than electricity or heat use. This is easily illustrated when looking at the CO_2-Equivalents per 1 kg of acetone compared to 1 kg of ethanol or 1 kWh of electricity, as shown in (Table 3). From Table 3 it is possible to visualize that, in terms of the overall LCA assessment, added acetone or ethanol weigh more than added heat or electricity, with acetone being two times more burdensome than ethanol. Nevertheless, the use of solvent in the PLE options is high enough that even though ethanol is less burdensome the total GWP impact outweighs the acetone use in the SE options.

In this regard, it is also worth mentioning that the ethanol used for this assessment is of petrochemical origin. However, since the waste being treated is wine pomace, it is quite possible that a biorefinery treating this waste would also produce bioethanol. This is true for distilleries placed in Italy and France, which currently treat wine pomace in order to produce bioethanol, bioenergy and food additives, among others (Lempereur and Penavayre, 2014). Bio-sourced ethanol will incur different environmental impacts, which were not investigated in this study.

Furthermore, the TEA in this study considers the processing costs including the financing costs. The market price of the product, the extracted polyphenols, and the market volume are yet to be explored. Once a market price or price range is known, then CapEx and processing costs can be compared to the benefits, and profitability indicators, such as net present value and internal rate of return. A larger investment for more complex technology (PLE instead of SE) might be justified if the benefits are significantly larger e.g. a higher yield for PLE than in the present study.

The most competitive options based on all midpoint impacts (Fig S2) and TEA; SE-10, SE-5 and PLE-10, were analyzed further to see if there is burden shifting between environmental indicators and to derive single scores for the options.

3.4. The single score results

After applying RIF, weighting strings can be derived for the application of TOPSIS with a range of importance given to economic impact from 0– to 1000, of a sum of 1000 available points distributed in the weighting profile between economic weight and environmental weight (Table S4). The relative importance amongst environmental impacts can also be shown in a single string to improve transparency of the weighting

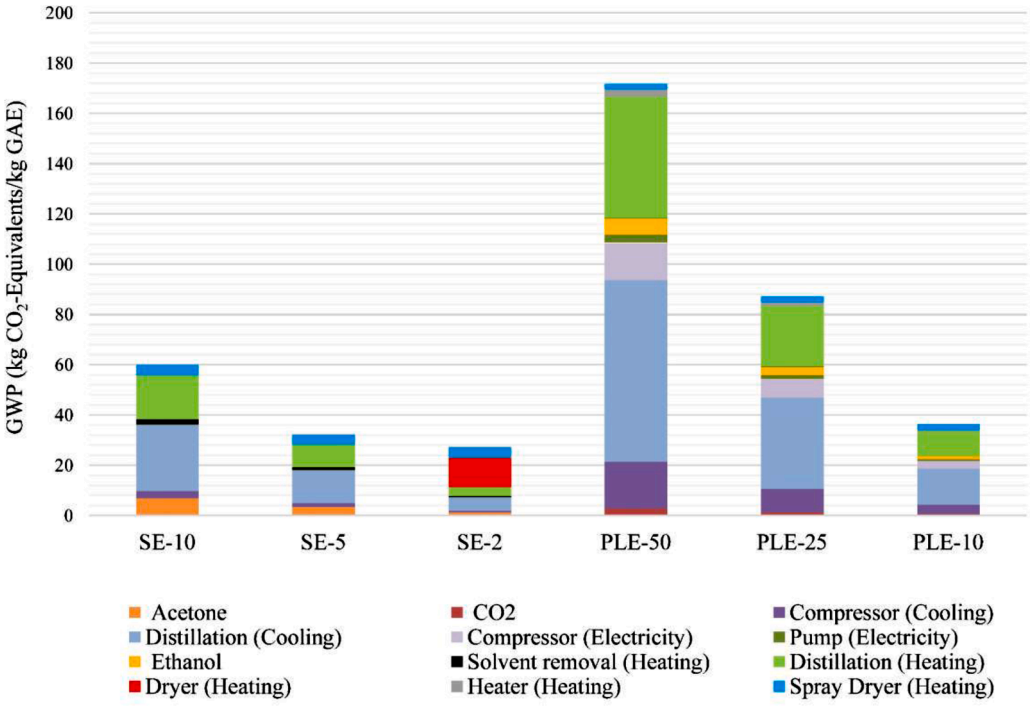

Fig. 5. Global warming potential for scenarios tested in kg of CO2-equivalents. Contribution per processing step, cut-off 1% of overall impact. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

(Table 4).

This is also done for equal weights (EW) amongst environmental impacts and the same range of importance of economic impact (Table S5). Applying these weightings to the criteria derived from LCA and TEA using TOPSIS, it is possible to provide decision support in the form of a single score indicator of idealness of the various technological alternatives including a relationship to environmental relevance across all impacts (Fig. 6). Furthermore, based on the results of the application of TOPSIS, a preference ranking can be made, with PLE-25 ranked fourth, SE-10 ranked third, and either PLE-10 or SE-5 ranked first and second. The ranking for first and second is based on the weight given to economics in the decision making process.

Based on the application of TOPSIS, it can be easily concluded that the PLE-10 and SE-5 methods outperform all other alternative extraction methods. While PLE-10 is the best economic performer, SE-5 proved to be the best environmental performer, though the differentiation between these is likely below the potential margin for error. This results in a shift in decision support depending on the weight given to economic factors, but again, this differentiation is likely not statistically significant. In addition, SE-10 consistently performs better than PLE-25 both environmentally and economically. This differentiation is statistically significant across all ranges of economic weighting. This results in a preference of SE-10 over PLE-25 regardless of weight given to economics. And, given that it is likely that an industrial process would be developed with a solvent ratio between the minimum and maximum solvent ratios as shown here for each technology respectively, it is apparent that there is more likelihood for SE to outperform PLE across all economic weightings (see SI figure S3).

As can be seen in Table 4 and Table 5, there is significant range in the importance of specific environmental impacts in RIF for the assessed methods. For example, some impacts such as human non-carcinogenic toxicity, marine eutrophication, and land use are insignificant in relative importance, and mineral resource scarcity is almost entirely irrelevant. On the other hand, fossil resource scarcity and freshwater ecotoxicity make up nearly half of weighting applied to environmental impacts due to the scale of their impact compared to the other environmental criteria relative to the average European's environmental impact.

One other element of note is the difference of decision support between EW and RIF in terms of the importance given to economic impact when PLE-10 is preferred over SE-5. When applying the RIF, this switch in preference occurs at appx. 65% weight to economic factors while for EW, the switch occurs at 55%. This is primarily due to the effective removal of environmental impact categories where the two alternatives are relatively equal that were compensating for other impact categories where the technologies were less equal in terms of performance. This occurs through the application of the ArgCW-LCA RIF weighting (Table 5) because some impact categories do not present much relevance to the decision context. This can be because there is either very little variation of the particular impact category amongst the assessed alternatives or because the given impact is smaller relative to status quo per capita emissions in relation to the other impacts of the assessed system.

Table 4
Weighting strings for RIF of environmental impacts used in this study, developed as described in Section 2.6.

Impact category	RIF	Impact category	RIF
Fine particulate matter formation	12.14	Marine ecotoxicity	171.22
Fossil resource scarcity	256.66	Marine eutrophication	0.94
Freshwater ecotoxicity	197.95	Mineral resource scarcity	0.004
Freshwater eutrophication	90.31	Ozone formation, Human health	22.35
Global warming	54.50	Ozone formation, Terrestrial ecosystems	26.86
Human carcinogenic toxicity	60.66	Stratospheric ozone depletion	2.06
Human non-carcinogenic toxicity	4.21	Terrestrial acidification	19.86
Ionizing radiation	31.02	Terrestrial ecotoxicity	39.87
Land use	0.60	Water consumption	8.79

Another important element in interpreting the results from RIF weighting is understanding that there is a level of uncertainty in the normalization factors used to derive the RIF, and that the decision to use current emissions as a reference point, i.e. by using a European's environmental impact as NF, does not necessarily have a relationship to the severity or consequences of environmental impacts. However, it does provide an indication of the relative importance of an emission, or reduction thereof, to the status quo. If absolute sustainability related factors were available for all relevant impact categories, the application of these instead of normalization factors would be preferable, as they would provide a stronger link to environmental impact. Ideally, this process would be completed relative to planetary boundaries (Steffen et al., 2015) using an absolute relationship to impacts from LCA (Bjørn et al., 2015). However, this cannot be done because this absolute relationship is not yet well enough understood/developed, nor has it been developed to include all impact categories covered in LCA.

An alternative to either of these methods would be to derive a RIF weighting from endpoints using e.g. monetization. While this might seem appealing, as there is a stronger connection with environmental damages when using endpoint indicators in LCA, the challenge comes in determining the relative importance of the different damage categories. This relative importance is purely subjective, and as such a specific cultural perspective would be applied to the derivation of the weighting profile. While this could be carried out in a scientific fashion to be representative of a decision maker group, the results would already contain some bias toward certain impacts introduced in the endpoint calculation (Kalbar et al., 2016; Sohn et al., 2017). This would make the results more challenging to interpret and potentially lead to decision support that in the end does not reflect the true preferences of the decision maker. And, though midpoint impacts are not devoid of subjectivity, utilizing RIFs based on midpoint impacts effectively reduces the layers of interpretation applied in the interpretation phase of the impact assessment relative to endpoint derived single scores. Thus, making the elements driving decision support easier to track and understand.

4. Conclusions

Polyphenol extraction methods were assessed using LCA at laboratory scale and a combination of TEA and LCA for designed industrial scale processes. Solvent to DW ratio and extraction yield are important parameters considering the design of the industrial scale processes, and therefore have a large impact on the results of the TEA and LCA. Thus, it is recommended that these parameters are optimized in the laboratory to ease their translation into industrial scale processes.

Out of the solvent to DW ratio ranges of the TEA-LCA, SE options have potential to perform better than PLE. Despite higher yields for PLE, higher economic and environmental burdens outweigh the benefit of higher yield for this option. The most important parameters indicated by the TEA are the polyphenol extraction yield and the solvent to DW ratio. The most important parameter for optimization indicated by the LCA results is reducing solvent amounts. The CFP at laboratory scale was useful in pointing out potential environmental hotspots, which served to guide the design of the industrial scale processes, from both an economic and environmental perspective. The single score indicator concluded that the potential performance is better when utilizing SE-5 than PLE-10, though a shift in preference is seen for higher economic weight. The addition of a transparent and reproducible decision assessment process aided in the understanding of the holistic impacts of the alternatives And, it can be concluded that the introduction of RIF as a method of deriving a weighting, relative to equal weights, for use in MCDA for LCA can likely reduce the impact of irrelevant and/or subjective criteria on the conclusions drawn from the application of MCDA that include

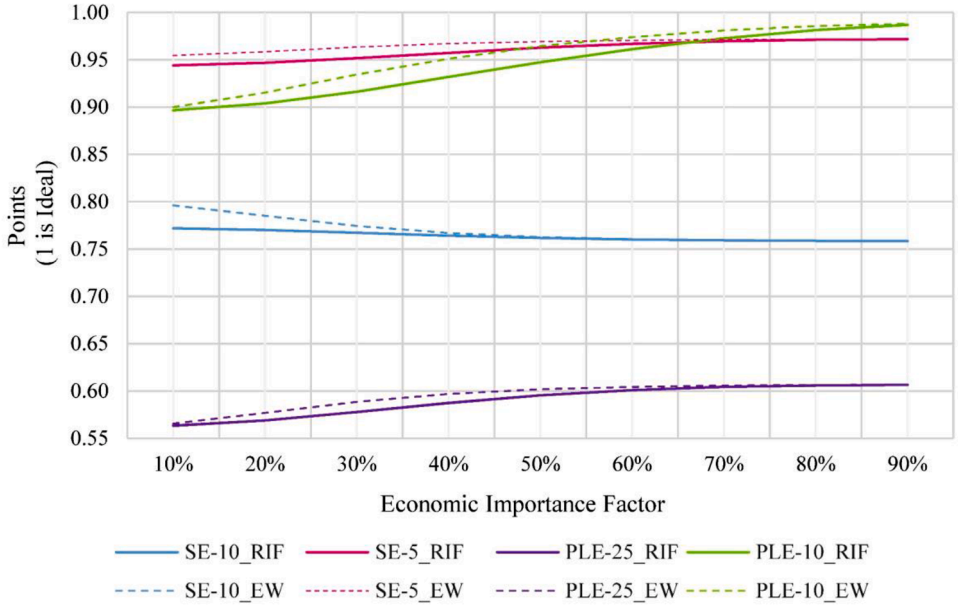

Fig. 6. TOPSIS derived single score indicator of idealness (most ideal=1) for both Relative Importance Factor (RIF) derived environmental weighting and Equal Weights (EW) environmental weighting amongst a range of weights given to economic performance. SE is solvent extraction, while PLE is pressurized liquid extraction. The number in each scenario indicates the solvent to DW ratio for the extraction process.

Table 5
Relative weight (RW) of environmental impacts between RIF and EW weighting ($\mathbf{RW} = \mathbf{W_{RIF}}/\mathbf{W_{EW}}$))

Impact category	RW	Impact category	RW
Fine particulate matter formation	21.85%	Marine ecotoxicity	308.19%
Fossil resource scarcity	461.99%	Marine eutrophication	1.69%
Freshwater ecotoxicity	356.31%	Mineral resource scarcity	0.01%
Freshwater eutrophication	162.56%	Ozone formation, Human health	40.23%
Global warming	98.10%	Ozone formation, Terrestrial ecosystems	48.35%
Human carcinogenic toxicity	109.18%	Stratospheric ozone depletion	3.70%
Human non-carcinogenic toxicity	7.57%	Terrestrial acidification	35.76%
Ionizing radiation	55.84%	Terrestrial ecotoxicity	71.77%
Land use	1.07%	Water consumption	15.83%

weighting such as TOPSIS. Furthermore, based on the application of TOPSIS, assuming that PLE-25 and SE-10 represent presently attainable solvent to DW ratios, while PLE-10 and SE-5 represent future potentially attainable solvent to DW ratios, it can be concluded that there is greater potential for better performance utilizing solvent extraction than pressurized liquid extraction across all value scales relating the environment and economics.

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.resconrec.2020.105318.

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