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Sustainability and circularity assessment of the potential of a biofuel produced from black liquor as a substitute for conventional fuels

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ABSTRACT

The European Bioeconomy Strategy aims to accelerate the deployment of a sustainable European bioeconomy to maximize its contribution to the 2030 Agenda and its Sustainable Development Goals (SDGs), as well as to the Paris Agreement on climate change. In this context, transport is considered as a key sector, with aviation and shipping playing an important role due to the need to meet its huge demand. In order to reduce potential emissions from the transport sector, the use of biofuels could be considered as a solution. However, with the aim of using biofuels to replace conventional fuels, it is important to assess their cost-effectiveness and feasibility, and in this aspect the valorization of waste streams for their production could help to meet this challenge. This is the framework of this research report, which is based on the use of black liquor (BL) from pulp and paper production to produce biofuel through a hydrothermal liquefaction (HTL) unit. Three technological designs and production capacities were considered: full extraction of the oil phase and its upgrading by catalytic hydrothermal deoxygenation (Scenario 1, S1), partial extraction of the oil phase (Scenario 2, S2) and full extraction but without upgrading (Scenario 3, S3). Low (100 t/d), medium (300 t/d) and high (600 t/d) production capacities were regarded.

From the modelling data, a life cycle perspective was adopted, taking into account both the environmental analysis (LCA) and the life cycle costs (LCC), as well as the circular potential using different performance, resource-flow circularity and economic indicators. In addition, a composite indicator, $CI_{LCA-LCC-CA}$, has been proposed to obtain a single score taking into account the three assessments: environmental, cost and circularity. The results obtained show that S3, with a production of 300 t/d, has the lowest environmental impact and the highest profitability corresponds to a capacity of 600 t/d. In terms of circularity, S3 also shows the best performance, mainly due to its higher resource productivity and lower energy intensity. These results are in line with those obtained for the composite indicator, and also show that higher production capacities and a simple but efficient process technology, such as S3, is the alternative with the highest potential for both sustainability and circularity.

1. Introduction

The widespread use of fossil fuels implies consumption of non-renewable resources as well as environmental impacts, which poses a challenge for long-term sustainability [29]. On the other hand, high demand for fossil fuels is subject to vulnerability issues associated with geopolitical instability, supply disruptions and supply chain fluctuations [25,31]. Energy diversification is necessary to ensure its availability and promote the transition to renewable energies, which have proven to be less harmful compared to energy based on conventional fuels, and also suitable for strengthening a circular bioeconomy Arias et al. [4,40,50].

In this sense, biofuels could be considered as a potential alternative for partial or total substitution of fossil fuels. The use of renewable fuels in the transport sector has risen from 12,107 thousand tons of oil equivalent in 2013 to 20,513 thousand tons of oil equivalent in 2022, according to the European database [16]. In relation to the shipping and

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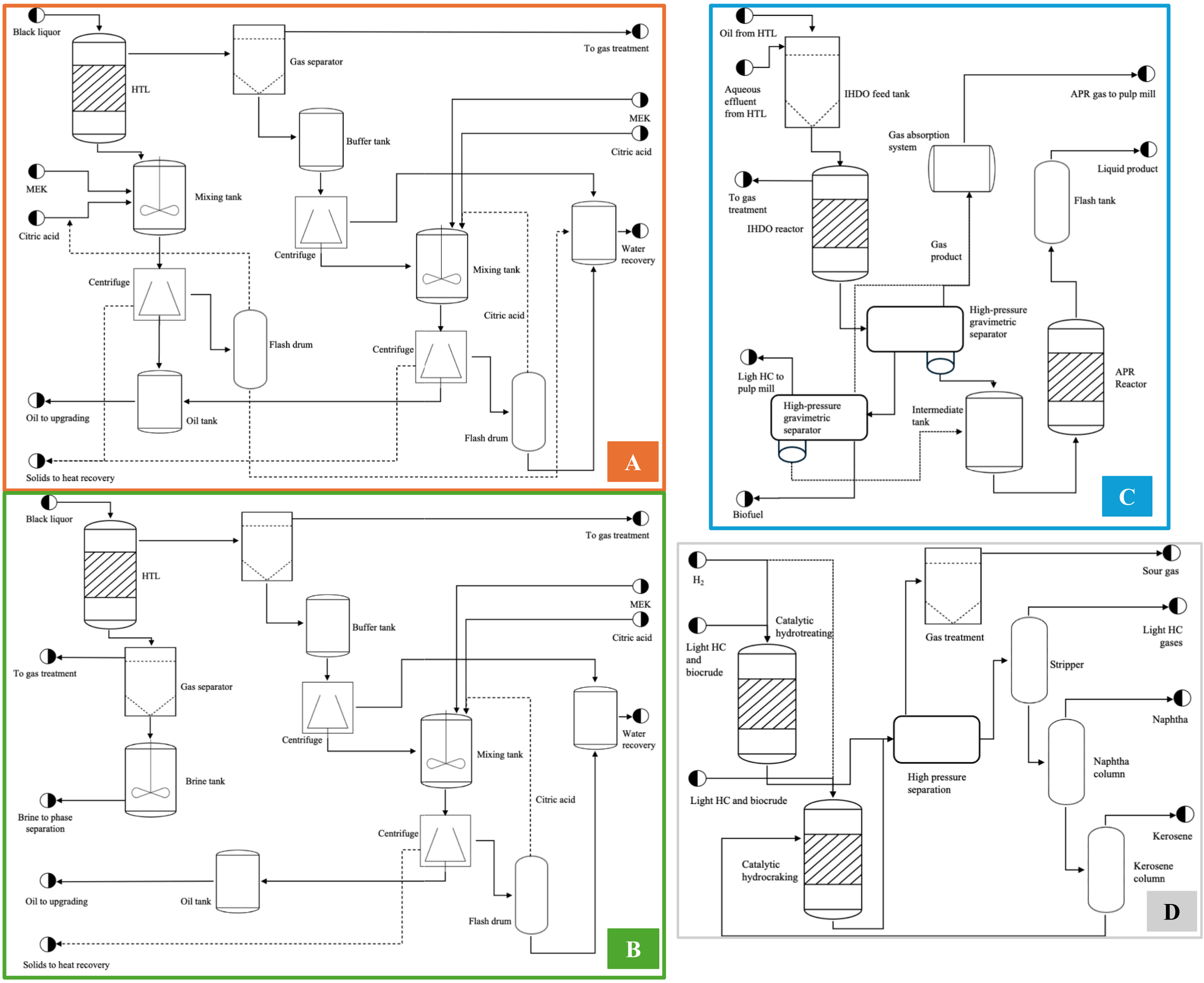

Fig. 1. Process diagrams: A represents S1 and S3, B represents S2, C is an intermediate stage required for S1 and S2 and D is the upgrading of the biofuels.

aviation sectors, the targets for the renewable fuels in shipping and aviation should be 19 % and 11 % respectively in 2030, rising to 85 % and 70 % in 2050 [22].

However, to promote the widespread use of biofuels, it is necessary to demonstrate their economic viability, their potential to reduce or mitigate environmental impacts, and to ensure that their development does not have other negative impacts on value chains. For example, 1st generation biofuels from food crops, an impact on the food supply chain could happen, as well as extensive use of land that could be used for agriculture, facing the controversy between the choice of energy supply or the food sector [20,41].

Due to this problem, alternative ways of biofuel production from the resources that cannot be used for food and feed appears as a better option [36]. In this approach, the use of waste streams appears to be the most viable alternative, although the treatment and processing required to use these wastes as feedstocks to produce biofuels is not necessarily simple and may require high-cost technologies or even involve high emissions that impact environmental quality [1,39]. Therefore, it is important to evaluate which residue is the most suitable for biofuel production, what type of technology is required, what production capacity is the most suitable to promote economic viability and, in addition, what is the profit margin and the potential of introducing it into the value chain, because if biofuels have a much higher production cost compared to fossil fuels, then their viability in the market will not be significant (Nair et al., 2022; [55].

With this in mind, the development of new biofuel production strategies should be considered as a key factor in the coming years, combined with a thorough and comprehensive assessment of their sustainability and market penetration potential. Specifically, in this research article, biofuels for shipping and aviation are produced using black liquor (BL) from the pulp and paper industry [48,54]. The main valorization of this stream is direct combustion for energy production, a process that not only supports internal operations, but also allows the sale of energy to the grid [12,37]. By allocating a percentage of the black liquor to biofuel production, the strategy ensures efficient use of this by-product, while the remaining percentage of black liquor continues to play a key role in the facility's energy management as a source of on-site power generation. This approach significantly reduces dependence on environmentally polluting fossil fuels, in addition to providing a high value-added product, improving both the environmental footprint and the profitability of the facility operations [27,33]. On the other hand, it is necessary to effectively assess whether these biofuels really imply an environmental, economic and circular benefit, for which it is essential to develop appropriate assessments, such as life cycle thinking [24,58], the use of circularity indicators [32,61], and also combining circularity and sustainability, using composite indicators [6].

The objective of this article is to assess two different type of biofuels, namely sustainable shipping and aviation fuels (SAF) (concretely bio-kerosene and bio-naphtha), which are required in order to fulfill the objectives of reaching, at least, a 5 % of SAF by 2030 in all flights departing from EU airports perform, using a comprehensive assessment encompassing environmental sustainability, economic viability, eco-efficiency and circularity performance. This article also develops a composite indicator, willing to be effective in disseminating results and could be considered as a tool for decision making on biofuel production schemes and it can contribute to the progress of biofuel production in the framework of a more resilient, sustainable and circular energy sector.It is hoped that the results achieved by this research could help stakeholders, entrepreneurs and policy makers on moving forward on the production of biofuels by waste, rather than opting for first generation biofuels, thus using edible crops, interfering on the food value chain.

2. Scenarios definition: Technologies and production capacities

Three scenarios were developed to provide technological performance alternatives considering two main process steps: biocrude production within a hydrothermal liquefaction (HTL) unit and its upgrading to biofuel. These scenarios were evaluated at three production capacities to determine the most favorable option, namely 100 t/d, 300 t/d and 600 t/d of BL valorization. It should be noted that the mentioned capacities of 100 t/d, 300 t/d and 600 t/d represent the valorization of about 5 %, 15 % and 30 % of BL, respectively, in order to maintain the circularity of the facility considering the production of energy for self-sufficiency by BL combustion. Regarding technological performances, S1 involves the production of biocrude with simultaneous separation of salts and hydrothermal liquefaction, leading to a complete extraction of the oily phase (Fig. 1). This phase is then upgraded using a catalytic hydrothermal deoxygenation (IHDO) process, alongside the production of hydrogen via Aqueous Phase Reforming (APR) (Fig. 1). This scenario emphasizes full utilization of both the brine and the HTL desalinated product. S2 corresponds the biocrude production process step of S1, but only extracting the oily phase from the HTL desalinated product. The brine is cooled, depressurized and directed to the pulp mill boiler. S2 focuses on partial upgrading of the extracted oily phase. Finally, in S3 the biocrude production process occurs in a single reactor, with complete extraction of the oily phase from both the salt brine and the HTL desalinated product. However, S3 does not involve hydrodeoxygenation or APR, but focuses on the extraction process considering an integrated hydropyrolysis and hydroconversion.

The rationales about using various technological designs and production capacities are: (1) the analysis of different technological schemes it could be identify the most effective approach for the maximization of production yield and biofuel quality, (2) by assessing various options of production capacities, it could be selected which of them is most suitable and optimal for industrial implementation, achieving an appropriate balance between production costs and biofuel economic benefits, and enhancing biofuel's long-term relevance and market competitiveness, (3) the comprehensive assessment of various technologies and production capacities could be helpful on the identification of the most sustainable methods for biofuel production, aligning with sustainability goals, European bioeconomy, standards and regulatory requirements, (4) as the amount of black liquor produced could vary between different geographical regions, assessing various production capacities could be helpful to identify the applicability of the production technologies and (5) deep analysis of technologies and production capacities could enhance the development of low-risk options for industrial-scale biofuel production, helping decision-making processes.

Table 1
Impact categories for assessing the environmental profile, and its normalization factors.

Acronym	Impact category	Units	Normalization factor
GWP	Global warming potential	kg CO_2 eq.	$1.25 \cdot 10^{-4}$
SOD	Stratospheric ozone depletion	kg CFC^{-1}1 eq.	16.7
TA	Terrestrial acidification	kg SO_2 eq.	$2.44 \cdot 10^{-2}$
FE	Freshwater eutrophication	kg P eq.	1.54
ME	Marine eutrophication	kg N eq.	$2.17 \cdot 10^{-1}$
TET	Terrestrial ecotoxicity	kg 1.4-DCB eq.	$6.58 \cdot 10^{-5}$
FET	Freshwater ecotoxicity	kg 1.4-DCB eq.	$3.97 \cdot 10^{-2}$
MET	Marine ecotoxicity	kg 1.4-DCB eq.	$2.30 \cdot 10^{-2}$
HCT	Human carcinogenic toxicity	kg 1.4-DCB eq.	$9.71 \cdot 10^{-2}$
HCNT	Human non-carcinogenic toxicity	kg 1.4-DCB eq.	$3.20 \cdot 10^{-5}$
FRS	Fossil resource scarcity	kg oil eq.	$1.02 \cdot 10^{-3}$
MRS	Mineral resource scarcity	kg Cu eq.	$8.33 \cdot 10^{-6}$

3. Methodology

3.1. Life cycle analysis (LCA)

3.1.1. Environmental LCA

Life Cycle Assessment (LCA) is an established methodology for quantifying the environmental impacts associated with all stages of a system or product life cycle, from raw material extraction to material processing, manufacturing, distribution, use, repair and maintenance, and disposal or recycling. This comprehensive framework allows to assess the environmental damage potential of the evaluated scenario, to identify the main contributors on the profile, i.e. the hotspots, to develop sensitivity assessments that allow to improve the sustainability potential of the scenario under evaluation and could also be considered as a guide for decision making [11,42,45]. It is standardized in ISO 14040, which provides guidance to enhance the effective use of this methodology, involving four main stages: definition of objectives and scope, life cycle inventory (LCI) analysis, life cycle impact assessment (LCIA) and interpretation of results. Each stage contributes to a holistic understanding of the environmental footprint of a product or process [2,3,8], as it provides a systematic perspective covering the entire life cycle, from raw material extraction to end-of-line. In this article, cradle-to-gate approach has been considered, from feedstock extraction to biofuels production, while as functional unit (FU) it has been the production of 1 kg of biofuel. ReCiPe 2016 Midpoint (H) V1.07 / World (2010) H has been selected as the calculation method, as it is one of the most widespread in the European context. The impact categories, along with the units of measurement and normalization factors, which are necessary to score the composite indicator, are depicted in Table 1.

3.1.2. Life cycle Costing (LCC)

Various industrial sectors have strived to gain competitive advantage by integrating both techno-economic and environmental aspects in their strategic planning [46]. This dual focus on process optimization, cost reduction and minimization of environmental impacts enhances both competitiveness and sustainability equally [18]. In this regard, Life Cycle Costing (LCC) method provides insights into the economic dimensions of sustainability, helping to formulate strategies that are economically viable [51]. The LCC concept, which emerged along with LCA in the mid-1970 s, helps decision makers improve the economic performance of a system life cycle, including accounting for costs associated with environmental externalities [21]; Fallah et al., 2013). This broad perspective is crucial for strategic business and policy decision making, as it provides a more detailed understanding of the

Table 2
Economic parameters selected for the analysis of the economic feasibility of the scenarios.

Acronym	Meaning	Formula	Units; Parameters meaning
CAPEX	Capital expenditure	Capital required to purchase goods and services	$
OPEX	Operational expenditure	Production costs	$
CE	Cost efficiency	$CE = \frac{OPEX}{CAPEX + OPEX}$	–
NPV	Net Present Value	$NPV = \sum_{t=1}^{T} \frac{C_t}{(1+r)^t} - C_0$	$; C_t net cash inflow at period t, C_0 total initial investment costs, r discount rate, and t time periods.
IRR	Internal Rate of Return	$0 = NPV = \sum_{t=1}^{T} \frac{C_t}{(1+IRR)^t} - C_0$	%; IRR is the discount rate for which the NPV is 0.
IE	Investment efficiency	$IE = \frac{NetPresentValue}{Capitalinvestment}$	–
PP	Payback peridod	$PP = \frac{\text{Initial Investment}}{\text{Annual Cash flow}}$	Years
A-NPV	Annualized NPV	$A\text{-}NPV = \frac{\text{NPV project}}{\text{NPV year i}}$	$

economic viability of a project.

The methodological steps of the LCC are similar to those used in LCA assessments and ensure that the LCC analysis provides a comprehensive and systematic assessment of the economics of proposed biofuel production scenarios. In LCC the required data include initial capital costs (e.g., equipment acquisition and installation, taxes, fees) and operating costs (e.g., energy, water, feedstock, consumable s, labor, maintenance). In this scenario, end-of-life costs (e.g., decommissioning, disposal, recycling) are not considered, as they are outside the scope.

Once all required cost data are collected, economic indicators are used to analyze the degree of economic profitability and the suitability of the evaluated scenarios. Various economic indicators are commonly used to evaluate and compare the financial performance of different options throughout their life cycles, the ones used for the assessment are described in Table 2.

3.1.3. *Eco-efficiency analysis*

Eco-efficiency, as defined by the World Business Council for Sustainable Development (WBCSD), further emphasizes the importance of providing quality goods and services while reducing ecological impacts and resource intensity (WBCSD, 1992). This concept has become a cornerstone for assessing sustainable development, advocating the reduction of resource consumption and environmental impacts without compromising product value [38]. In fact, the eco-efficiency score is accomplished through three main objectives: increasing product quality, optimizing resource use and reducing environmental burdens (Cucek et al., 2015). Eco-efficiency analysis, taking advantage of the synergies between LCA and LCC, provides a quantifiable measure of sustainability, thus aligning environmental and economic objectives.

The eco-efficiency analysis proposed in this report has been based on the consideration of the most relevant environmental impact categories, namely global warming potential (GWP) and fossil resource depletion (FRS), given their importance and frequent evaluation in the assessment of the environmental impacts of (bio)fuels. And, as for the economic dimension, Capital Expenditures (CAPEX) and Operating Expenses (OPEX), chosen for their independence from variable revenues in different scenarios. Taking into account this economic approach, a more equitable comparison is developed between all the scenarios considered, providing a clear picture of the costs involved without the variability introduced by the different revenue streams.

For the representation of the eco-efficiency results, diagrams have been used, in which an economic indicator versus an environmental indicator, resulting in four distinct area-plots. Each plot-area visually delimits the eco-efficiency regions: the area shaded in red marks the non-eco-efficient zone, green signifies the eco-efficient zone and yellow indicates an intermediate zone, all demarcated by the eco-efficiency line. This graphical representation serves as a valuable tool to identify which scenarios offer a balance between environmental sustainability and economic viability, guiding the selection of the most eco-efficient technology and capacity.

3.1.4. *Circularity assessment*

The Circular Economy (CE) emphasizes the transition from linear production and economies, characterized by the resource depletion and environmental degradation of the process-consume-dispose model, to the promotion of closed-loop systems. In these systems, raw materials and waste are recovered and valorized [49,28,7]. This shift aims to extend the life cycle of products and materials, minimize waste generation, reduce environmental damage, conserve resources and improve economic resilienceVelenturf and Purnell [60,17]. The EC strives to close the gap between resource demand and availability, considering waste as a potential resource to foster sustainable and waste-free value chains [30]. Within this paradigm, biorefineries and biotechnological processes emerge as viable avenues for sustainable and circular production by improving renewable resource utilization, process efficiency and waste reductionArias et al. [5,13].

The main aspects and principles included in a circular economy strategy are: resource efficiency, creating more value (both in terms of production capacity, economic and social aspects) with less resource input [26]; waste prevention: reducing the amount of waste generated by redesigning the production process or product [53]; waste recovery and/or resource recovery and/or recycling: considering the reuse or recovery of waste for the production of high value-added products [57];

Table 3
Circularity indicators considered for scoring the circularity performance of the scenarios.

Acronym	Type	Formula	Units
BOP[1]	P	Bio-oil production: $\frac{\text{kg of produced bio oil}}{\text{kg of black liquor used}}$	kg/kg
ERI[1]	P	Energy Return on Investment (bio-oil): $\frac{\text{Gross energy produced (MJ)}}{\text{Local/Upstream energy inputs (kWh)}}$	MJ/kWh
CPFI[1]	C	Circular process feedstock intensity: $\frac{\text{Raw materials (t)}}{\text{product + coproducts + recovered material (t)}}$	t/t
RE[1]	C	Renewable energy [%]: $\frac{\text{Renewable energy used or produced (kWh)}}{\text{Total energy consumption (kWh)}}\hat{A}\cdot 100$	kWh/kWh
CPEI[1]	C	Circular Process Energy Intensity: $\frac{\text{Fossil + renewable - internal derived energy (kWh)}}{\text{mass product + co-product + recovered products (kg)}}$	kWh/kg
RP[1]	C	Resource productivity: $\frac{Total\ sales\ (M\$/year)}{Virgin\ material\ inflow\ (kg/year)}$	M$/kg

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[1] Acronyms: BOP (Bio-oil production), ERI (Energy Return on Investment), CPFI (Circular Process Feedstock Intensity), RE (Renewable Energy), CPEI (Circular Process Energy Intensity), RP (Resource Productivity), P (Performance indicator), C (Circular Resource flow indicator).

Table 4
Characterization impact results per impact category for the scenarios considering a production capacity of 100, 300 and 600 t/d of BL. [1]Note: all data is represented per functional unit (1 kg of bio-oil produced).

Acronym	Units	100 t/d[1]			300 t/d[1]			600 t/d[1]		
		S1	S2	S3	S1	S2	S3	S1	S2	S3
GWP	kg CO_2 eq.	35.09	34.56	30.93	35.00	34.53	30.85	35.03	38.38	30.88
SOD	kg $CFS1_1$ eq.	$1.47{\cdot}10^{-5}$	$1.37{\cdot}10^{-5}$	$1.30{\cdot}10^{-5}$	$1.47{\cdot}10^{-5}$	$1.36{\cdot}10^{-5}$	$1.30{\cdot}10^{-5}$	$1.47{\cdot}10^{-5}$	$1.52{\cdot}10^{-5}$	$1.30{\cdot}10^{-5}$
TA	kg SO_2 eq	0.12	0.12	0.11	0.12	0.12	0.11	0.12	0.13	0.11
FE	kg P eq	0.03	0.03	0.02	0.03	0.03	0.02	0.03	0.03	0.02
ME	kg N eq	$3.45{\cdot}10^{-3}$	$3.32{\cdot}10^{-3}$	$3.05{\cdot}10^{-3}$	$3.45{\cdot}10^{-3}$	$3.32{\cdot}10^{-3}$	$3.04{\cdot}10^{-3}$	$3.45{\cdot}10^{-3}$	$3.69{\cdot}10^{-3}$	$3.04{\cdot}10^{-3}$
TET	kg 1.4-DCB eq	59.84	56.68	52.83	59.78	56.66	52.78	59.80	62.94	52.80
FET	kg 1.4-DCB eq	0.35	0.32	0.31	0.35	0.32	0.31	0.35	0.36	0.31
MET	kg 1.4-DCB eq	0.50	0.46	0.44	0.50	0.46	0.44	0.50	0.51	0.44
HCT	kg 1.4-DCB eq	0.73	0.71	0.65	0.73	0.71	0.64	0.73	0.78	0.65
HCNT	kg 1.4-DCB eq	13.82	13.09	12.20	13.72	13.07	12.11	13.74	14.53	12.13
MRS	kg Cu eq	$1.39{\cdot}10^{-2}$	$1.17{\cdot}10^{-2}$	$1.22{\cdot}10^{-2}$	$1.38{\cdot}10^{-2}$	$1.17{\cdot}10^{-2}$	$1.22{\cdot}10^{-2}$	$1.38{\cdot}10^{-2}$	$1.30{\cdot}10^{-2}$	$1.22{\cdot}10^{-2}$
FRS	kg oil eq	11.58	11.47	10.22	11.56	11.46	10.20	11.57	12.73	10.20

and reuse and/or recycling of resources, considering the same use or an alternative use for a material or waste produced instead of managing it as waste [47].

Once the key aspects have been identified, a thorough analysis should be conducted to determine whether a scenario under analysis aligns with the principles of the circular economy. This involves quantifying all input and output flows, covering raw materials needed, volume of products, by-products and waste streams, as well as emissions. Furthermore, beyond these flows, the assessment of circularity must also examine the extent to which required materials are conserved through recycling, reuse or refurbishment. This includes assessing the estimation of the share of recycled materials relative to total waste produced, the amount of waste used as feedstock for energy generation, among others.

Another critical aspect of circularity assessment is the evaluation of energy efficiency, defined by the energy required per amount of product produced [34]. This involves taking into account energy inputs from the grid, from waste valorization, and also energy outputs, especially if the process can produce surplus energy that could be sold to the grid. In this context, the lower the energy required to produce a specific amount of product, the better the circularity score [19].

In order to evaluate all aforementioned elements the reports developed by circularity practitioners, available guidelines and reports have been used, as the Ellen MacArthur Foundation [35], the BS 8001 British Standard [9], the Circular Economy Action Plan,[56], the OECD Inventory of Circular Economy Indicators [44] and the European Union Monitoring Framework [52]. The final selection of indicators is depicted on Table 3.

3.2. Composite indicator

Additionally, as an innovative aspect and as a method of aggregation, the development of a composite indicator has been considered, which takes into account both the results derived from the environmental assessment following the LCA methodology, the economic scores ob-

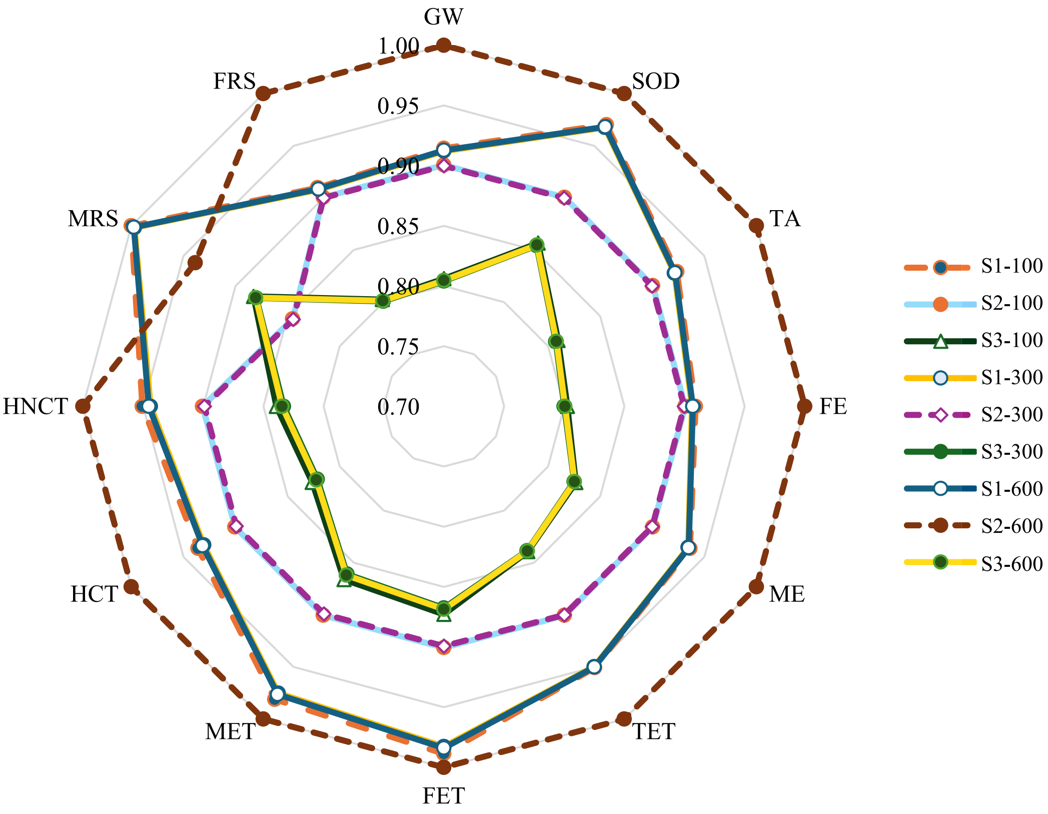

Fig. 2. Comparison between scenarios and production capacities.

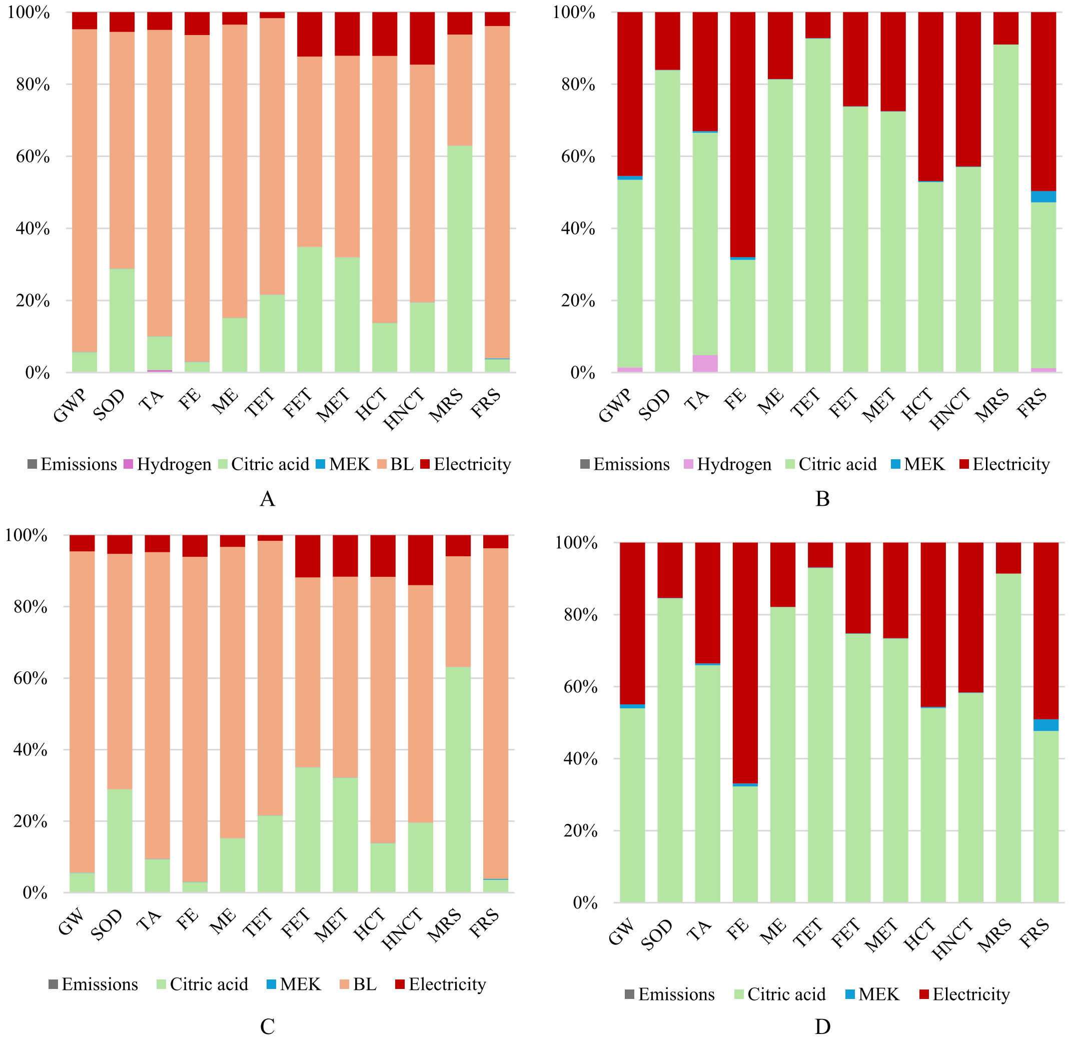

Fig. 3. Environmental profiles of worst- and best-Scenario scenario. A and B represents S2 – 600 t/d, while B considering black liquor as a zero-impact residue, C and D reports the same but for S3 – 300 t/d.

tained from the LCC and the values of the circularity indicators. Each of these pillars, environmental, economic and circular, has an equal weight in the final value of the indicator, using a factor of 1/3 for each parameter, as shown in Equation (1) (Eq. (1).

$$CI_{LCA-LCC-CA} = CI_{LCA}\hat{A}\cdot f_{CI} + CI_{LCC}\hat{A}\cdot f_{CI} + CI_{CA}\hat{A}\cdot f_{CI} \tag{1}$$

Where CI_{LCA} is the individual indicator referring to the environmental perspective following LCA methodology, CI_{LCC} is the one referred to the LCC and CI_{CA} the sub-indicator used to measure the circular perspective, while f_{CI} is the factor considered for the $CI_{LCA-LCC-CA}$ indicator, which is 1/3.

To calculate the value of the individual indicator associated with the environmental perspective, it was necessary to normalize the environmental impact values obtained, using the normalization factors included in Table 1. In this way, the environmental scores can be dimensioned. On the other hand, in order for each of the categories, which are 12 in total, to have the same weight on the final value of the environmental sustainability indicator, a factor of 1/12 is applied to each of the normalized impact categories, as shown in Equation (2) (Eq. (2).

$$\begin{aligned} CI_{LCA} = {} & GWP\hat{A}\cdot f_{LCA} + SOD\hat{A}\cdot f_{LCA} + TA\hat{A}\cdot f_{LCA} + FE\hat{A}\cdot f_{LCA} + ME\hat{A}\cdot f_{LCA} \\ & + TET\hat{A}\cdot f_{LCA} + FET\hat{A}\cdot f_{LCA} + MET\hat{A}\cdot f_{LCA} + HCT\hat{A}\cdot f_{LCA} \\ & + HNCT\hat{A}\cdot f_{LCA} + MRS\hat{A}\cdot f_{LCA} + FRS\hat{A}\cdot f_{LCA} \end{aligned} \tag{2}$$

Where f_{LCA} is the factor considered for the CI_{LCA} indicator, which is 1/12.

Regarding the individual indicator associated with economic aspects, the most commonly used financial indicators for project evaluation, i.e., NPV, investment efficiency, and IRR, were considered. As previously, a weight factor of 1/3 is applied to maintain an equivalent weight among the economic factors considered (Equation (3), Eq.3).

$$CI_{LCC} = NPV\hat{A}\cdot f_{LCC} + IE\hat{A}\cdot f_{LCC} + IRR\hat{A}\cdot f_{LCC} \tag{3}$$

Where f_{LCC} is the factor considered for the CI_{LCC} indicator, which is 1/3.

Finally, with regard to the circularity indicator, the six indicators pre-selected in Table 3 have been considered, taking into account that two of them, specifically the CPFI and the CPEI require an inverse value since, the higher the value of these parameters, the lower their circular potential. The final equation for the circularity sub-indicator, considering again a weighting factor, is included in Equation (4) (Eq. (4).

$$CI_{CA} = BOP\hat{A}\cdot f_{CA} + ERI\hat{A}\cdot f_{CA} + \frac{1}{CPFI}\hat{A}\cdot f_{CA} + RE\hat{A}\cdot f_{CA} + \frac{1}{CPEI}\hat{A}\cdot f_{CA} + RP\hat{A}\cdot f_{CA} \tag{4}$$

Where f_{CA} is the factor considered for the CI_{CA} indicator, which is 1/6.

4. Results

4.1. Environmental profiles and impact categories burdens

The values obtained for each of the selected impact categories of the ReCiPe MidPoint calculation method are shown in Table 4, while the graphical representation of the global comparison is provided in Fig. 2. The highest impact values are highlighted in red in Table 4, while the lowest in green. Thus, S2, together with the highest production capacity, 600 t/d, is the one with the highest environmental damage values (Fig. 2), highlighting the FE, GWP, FRS and TA categories, with impact values of 25.0 %, 24.4 %, 24.9 % and 23.9 % higher than the scenario with the lowest impact value per category.

On average, S2 with 600 t/d of production capacity has an impact of 20.0 % higher in comparison, in this Scenario, with S3, with production capacity of 300 t/d, being this the best scenario, i.e., the one with the lowest environmental impact in all impact categories under study. The greater simplicity of the technological process, as well as the complete extraction of the oily phase, implies a higher production of bio-oil per batch, which has an important effect on the overall impact of the process.

In addition, this S3 also does not use hydrogen as an input, which, although it does not show a significant impact observing the environmental profile of S2 (Fig. 3A), does have some effect on the GWP, TA and FRS categories. On the other hand, another aspect to remark is that S3 does not require the aqueous phase reforming stage, thus avoiding the energy requirements of this step, as well as the emissions or waste streams associated with it.

On the contrary, S2 shows the worst results mainly because it is the technological model that shows a lower productivity in the production of biofuels, although it is the scenario that requires a lower input of citric acid, which implies an environmental benefit in the MRS category, as shown in Table 4, being this scenario the one that shows the best result, this benefit is not enough to counteract its lower production efficiency.

As for S1, it is in an intermediate position between S3 and S2, although it is closer to the results of S3. The highest percentages of impact increase with respect to S3 amount to 14.1 % for the AT impact category and 13.44 % for the GWP impact category, with an average value of 13.4 % increase, thus being practically half of the comparative values of S2.

An important aspect to evaluate, in addition to the type of technological model most suitable from the environmental point of view, is also the selection of the production capacity that contributes to reduce the environmental impacts per kg of biofuels produced. While for S1 there is a tendency to reduce or maintain the impact score by increasing the production capacity, for S2, by increasing the production capacity to 600 t/d, there is a noticeable increase in the environmental impact per kg of biofuel produced.

On the other hand, once the environmental impact values obtained have been evaluated, it is also important to analyze which elements of the inventory are contributing most notably to them, i.e., to identify the hotspots. Fig. 3 shows four environmental profiles, Fig. 3A corresponds to the worst-scenario, S2 – 300 t/d, same Scenario for Fig. 3B, but considering the BL as a waste with zero impact. The objective of performing this analysis is to identify what are the impacts of the process technology, which, as can be observed, citric acid is the main contributor in most of the impact categories, with the exception of the FE, FRS and GWP categories where electrical energy requirements also have an important weight. The same impact contributions are observed for Fig. 3C and Fig. 3D, both corresponding to S3 – 300 t/d, with the only exception of the absence of hydrogen impact, since it is not required in this technological process alternative. Hydrogen is used in hydrogenation step to upgrade bio-oil, but if it is not optimally integrated into the process may not fully use the biogenic carbon available in the black liquor, leading to incomplete conversion, thus lower yields and higher quantity of residual carbon-rich waste. And this also have an effect over the higher energy costs and increased carbon penalties, reducing the potentiality of the produced biofuels. That is why it is important to optimize the use of hydrogen to favor the higher production yield. Besides, looking to increase the renewability and sustainability potential of the biofuel production process, the use of green hydrogen not only avoid the reliant on non-renewable inputs, but also maximize the environmental benefits, which implies an improvement and enhancement of the biofuels production process indicators (as the ones of circularity or the ones related with greenhouse gases emissions, for example).

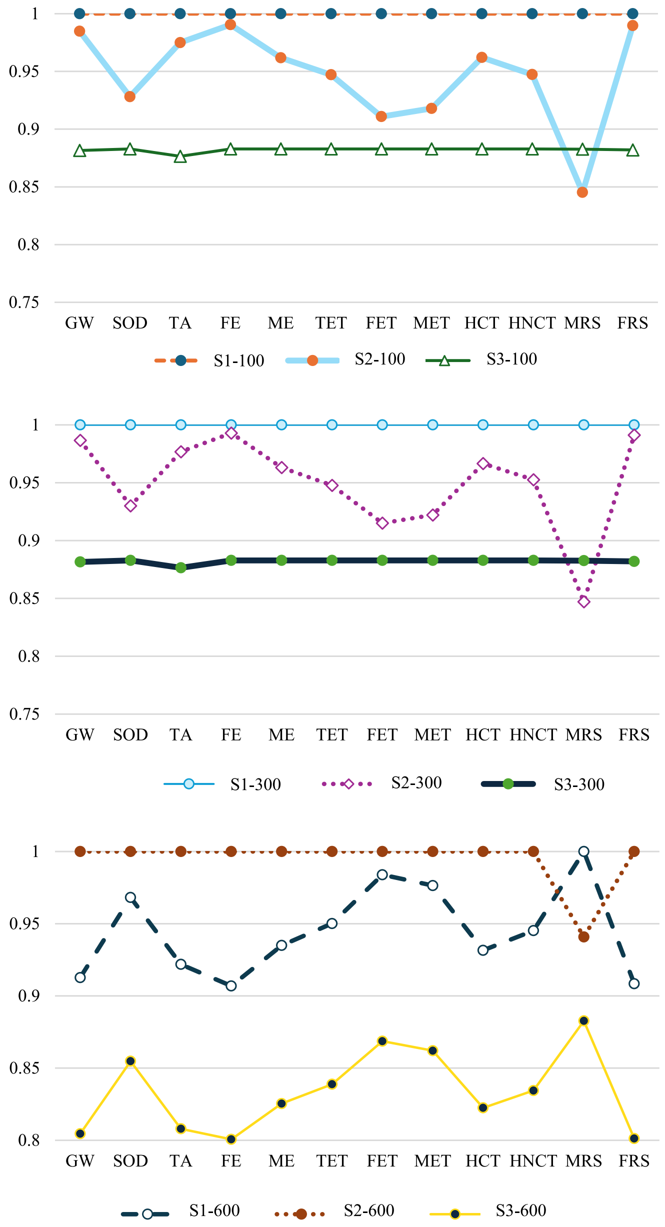

Fig. 4. Comparison between scenarios and production capacities.

As can be seen, the environmental profiles of both scenarios are very similar, implying that the better environmental performance of S3 with respect to S1 derives from its higher production capacity, rather than from chemical consumption, emissions associated with black liquor production or energy requirements.

4.1.1. Comparison between scenarios

In addition to an overall comparison, a comparison by production capacity is also included to determine the technological process model that contributes to a lower environmental impact (Fig. 4).

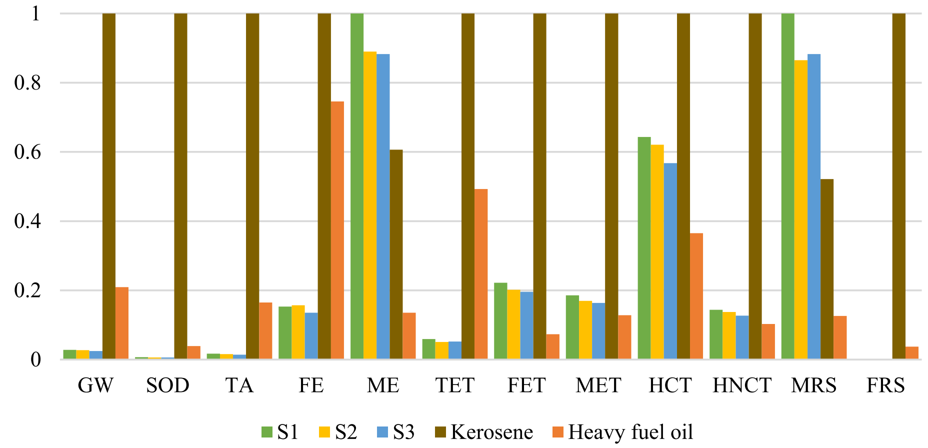

Fig. 5. Comparison with environmental loads of fossil fuels: kerosene and heavy fuel oil.

When 100 t/d is analyzed as the production capacity, S3 yields the highest amount of biofuel and also the one that implies the least environmental loads, notably because it bypasses the emissions typically associated with the aqueous phase reforming stage in its production model. The only exception is observed in the impact category of MRS, in which is S2 the one that exhibits the lowest impact value. This anomaly is mainly attributed to the energy demands of S3, which are higher than those of S2, needing a total of 2239.4 kWh, in contrast to the 2132.5 kWh required for S2.

S3 also stands out by producing the most bio-oil at 1.72 t/d when analyzing a production capacity of 300 t/d, which is more than both S1 and S2, and without requiring any hydrogen input, an element necessary in the other two cases. Moreover, S3 results in fewer emissions and waste requiring treatment, further distinguishing its process efficiency and environmental profile. While S1 also shows increased bio-oil production at 300 t/d, S2 sees a slight decrease. Energy requirements for S3 are similar to those in S1 and marginally higher than in S2. These factors collectively demonstrate that S3, with its higher bio-oil output and lower emissions, could offer better environmental performance at the 300 t/d production level compared to the other scenarios. When analyzed 600 t/d, S3 also exhibits the lowest environmental impact across all the impact categories, thus emerging as the unequivocal leader in environmental sustainability, outperforming the others in all impact categories. The second significant change concerns the scenario with the highest environmental impact. Unlike the previous analysis, S2 is now identified as having the highest environmental impact, marking it as the least sustainable option among the scenarios considered.

There is some analogy between production capacities, with S3 being the one that implies a lower environmental contribution in all of them, with the only exception of the MRS category, which, as mentioned above, the reduced load derives from the reduction of citric acid consumption of S2, for the low and medium production capacity (Fig. 4). However, for 600 t/d, in the same impact category, the environmental benefit for S2 is not noticed, being S3 the one that, with a significant difference, achieves the best results in all impact categories, highlighting those of GW, TA, FE and FRS, whose impact is reduced by about 20 % compared to S2, which is the one that implies the highest environmental contribution in this Scenario.

It should be noted that S2 has a higher environmental impact by increasing the production capacity from 300 t/d to 600 t/d. Its lower production yield compared to the other two scenarios seems to have a higher environmental impact when increasing production capacity. However, the opposite effect is observed for S1, which has the highest potential impact for both low (100 t/d) and medium (300 t/d) production capacities but is in an intermediate position when evaluated at 600 t/d (Fig. 4). Therefore, it can be concluded that the increase in production capacity is beneficial for the technological model of S1. The same is true for S3, since while the reduction in impact with respect to the highest contribution scenario is around 11.46 % for 100 t/d and 300 t/d, the reduction increases to 16.14 % when evaluating the 600 t/d capacity.

Once analyzed all production capacities and technologies under assessment, it could be underline that a 300 t/d capacity presents an optimal balance between environmental sustainability and operational feasibility. This capacity, representing 15 % of BL valorization, is in accordance with the identified sustainability threshold, avoiding negative impacts on the kraft pulp mill's environmental profile. The analysis further elucidates that while higher capacities offer increased bio-oil production, the marginal environmental benefits diminish, especially beyond the 15–20 % BL valorization mark. This finding is important, suggesting that efforts to scale beyond this point must be approached with caution to prevent undermining the environmental integrity of the associated pulp mills.

To assess the viability of the bio-oil produced within the value chain and fuel market, it is crucial to compare it with conventional fuels, specifically kerosene and heavy fuel oil, which are predominantly used

Table 5
LCC results. Acronyms[1]: PC (production capacity), MSP (Minimum Selling Price).

Acronym	Units	PS[1]	100 t/day			300 t/day			600 t/day		
		Scenario	S1	S2	S3	S1	S2	S3	S1	S2	S3
		MSP[1]	5.5	5.5	2.0	5	4.5	2.0	4.5	5.5	1.5
CAPEX[1]	M$		29.65	30.83	19.40	69.95	119.3	46.01	119.3	203.4	80.69
OPEX[1]	M$		−75.76	−51.88	−46.23	−214.9	−301.0	−171.6	−301.0	−180.1	−142.1
CE[1]	–		1.64	2.46	1.72	1.48	1.66	1.37	1.66	1.61	2.31
NPV[1]	M$		16.31	0.64	8.65	60.42	63.35	58.09	63.35	40.86	5.49
IRR[1]	%		19	12	18	24	19	29	19	20	13
IE[1]	–		0.52	0.02	0.45	0.86	0.53	1.26	0.53	0.35	0.07
PP[1]	Years		6	7	6	5	6	4	6	4	3
A-NPV[1]	M$		0.65	0.05	0.74	5.18	5.44	4.99	5.44	3.51	0.47

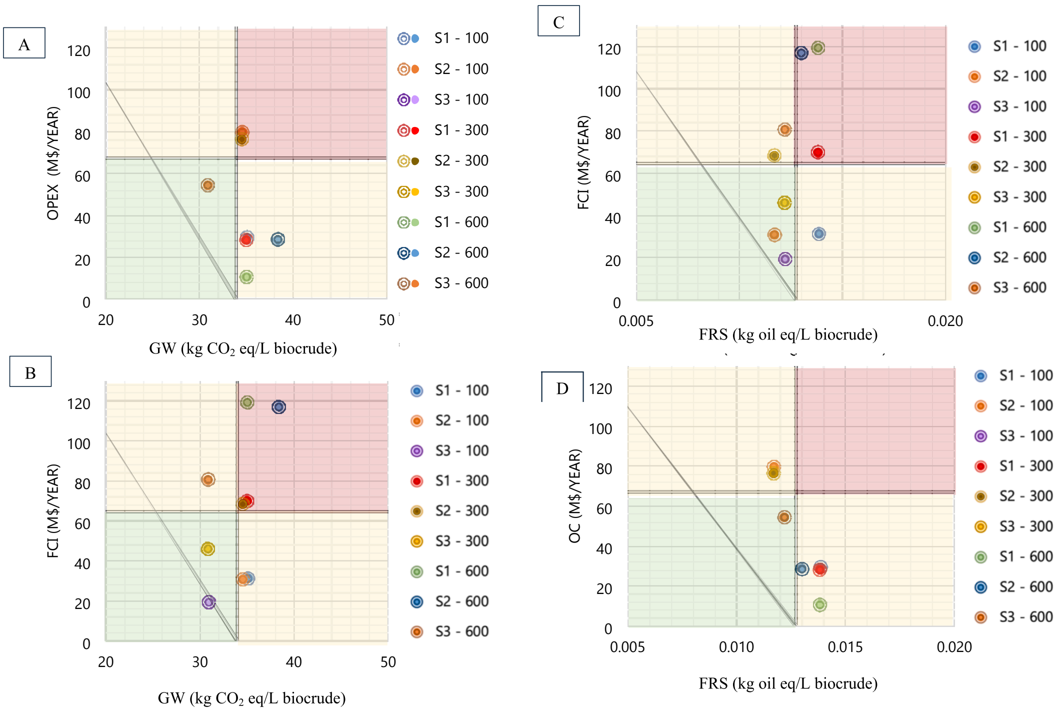

Fig. 6. Eco-efficiency analysis considering the comparison between CAPEX and OPEX as economic indicators and GW and FRS as environmental impact categories.

in the aviation and maritime industries. For this comparison, a production capacity of 300 t/d was chosen, using the same functional unit (1 kg of fuel produced), considering only the bio-oil production process, and using the same calculation methodology and impact categories for consistency. As illustrated in Fig. 5, fossil fuels exhibit significantly higher impacts in most categories, except for ME and MRS. The greatest burden on these impact categories stems from the use of chemicals in the process, with citric acid standing out in the environmental profile, and energy requirements, which are far from optimized compared to conventional fossil fuels. In terms of technology, in almost all impact categories evaluated, S3 achieves the most favorable environmental scores. This superiority is attributed to the complete extraction of the oily phase from both the brine and the HTL desalinated product, underscoring the environmental effectiveness of this approach in bio-oil production.

However, it is clear that, although the stage of development of this biooil is less advanced than that of fossil fuel production processes, biooil derived from black liquor valorization suggests greater environmental benefits. Since the production model is currently at pilot scale, there is great potential for improvement and optimization, which in turn could improve its environmental performance.

4.2. LCC results. Analyzing economic feasibility

A range of minimum selling prices (MSP) of bio-oil in $/kg has been considered, which, as can be seen in Table 5, ranges from a minimum of $1.5/kg to a maximum of $5.5/kg, with the MSP being the minimum selling price required to ensure the economic viability of the plant, i.e. for the net present value to be positive. In this regard, the production capacity, which is directly related to the profit-making capacity, as well as with the costs derived from the consumption of chemicals and energy, have a high impact on the economic viability of the plant. The lower the MSP required to achieve a positive NPV, the more profitable the process.

Comparing the CAPEX values, mainly derived from the equipment acquisition, it is observed that S3 has the lowest value, mostly because this process does not require the intermediate stage with the APR and IHDO reactors, Fig. 1C, while S1 and S2 do. On the other hand, in terms of OPEX, S3 remains the scenario with the lowest operating costs, and between Scenarios 1 and 2, it is S2 that has the lowest OPEX, due to the reduction in the consumption of citric acid in the separation stage, which represents 7 % of the total cost of consumables and utilities and thus has a notable effect on this parameter.

As for the EC parameter, since it represents efficiency, the higher the value, the better. In this Scenario, production capacity plays an important role in selecting the best-scenario. For low and medium production capacities, S2 is the most economically efficient, while when evaluating the 600 t/d capacity, it becomes the least efficient alternative, with the best scenario being S3. On the other hand, with respect to S1, it is classified as the least efficient scenario for 100 t/d, and the second most efficient for medium and high production capacities.

Regarding the other economic parameters studied, all of them depend on the selling price of the product obtained, therefore the value of the MSP that ensures the economic viability of the process must be considered. With a significant difference, S3 is the most efficient production model, since the minimum selling prices fall to values between 1.5 and 2 $/kg, while S1 and S2 require MSPs between 4.5 and 5.5 $/kg. For this reason, S3 is considered the most economically viable model, and it also shows robustness to price variations within the evaluated range. And this robustness is critical in ensuring that the economics of the process are not overly sensitive to price fluctuations and can maintain profitability even at the lower end of the price scale. In this sense, S3 could be considered the preferred choice for scalability and commercial implementation.

When comparing S1 and S2, which have similar MSP values, S2 would be preferred for medium production capacities, i.e. 300 t/d, while S1 is preferable for low and high production capacities, as it has a higher investment efficiency and a higher NPV.

4.3. Eco-efficiency analysis

This section includes the eco-efficiency results across various production capacities and Scenario scenarios. The intention is to harness these insights to enforce the reasoning for the selection of the most advantageous option, offering a dual lens to evaluate the sustainability of biocrude production from black liquor. This analysis aims to discern

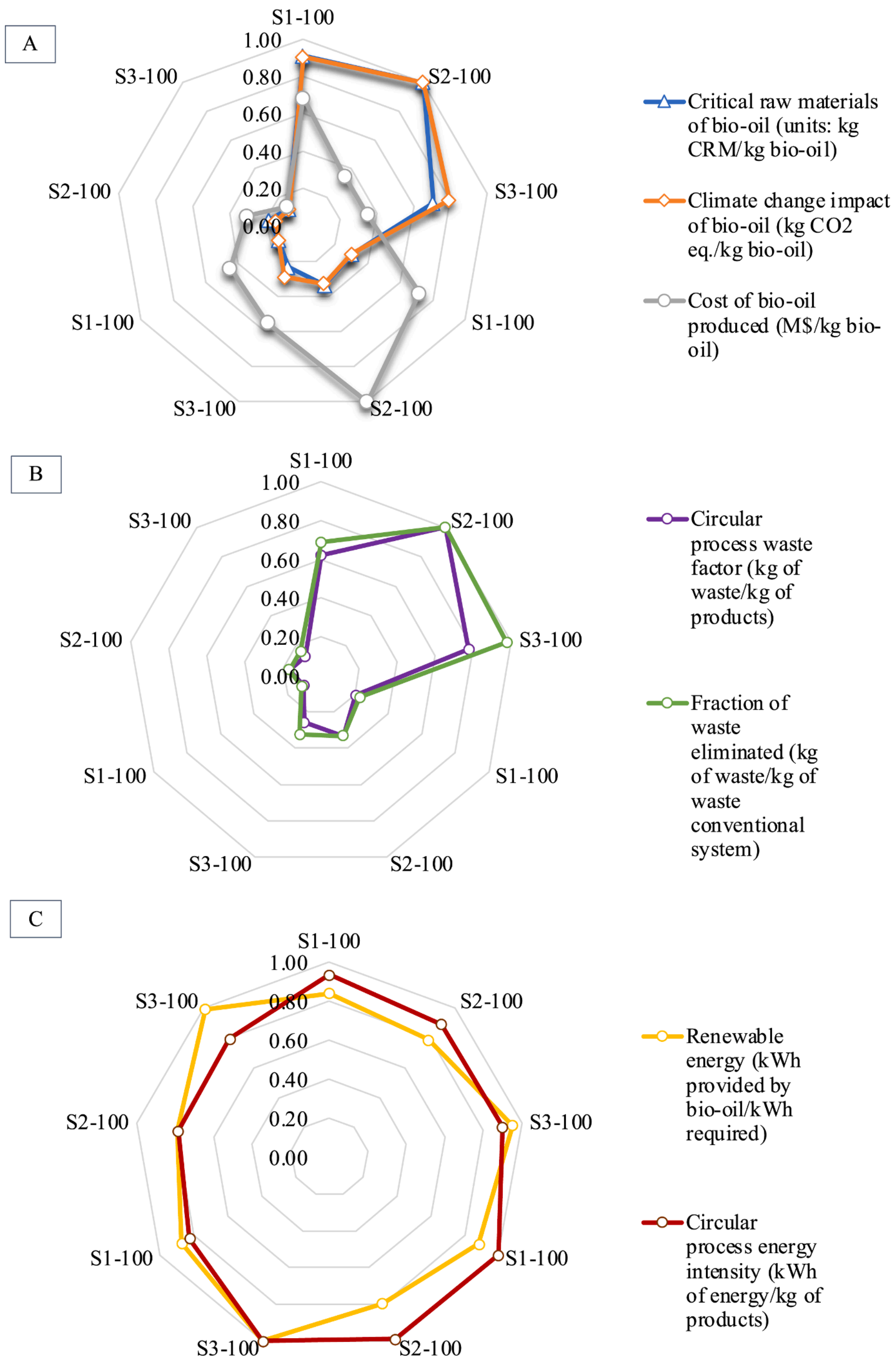

Fig. 7. Comparison between selected circularity indicators.

the optimal balance between minimizing ecological impacts and ensuring economic viability, paving the way for more informed, sustainable decision-making. The eco-efficiency results for the selected economic and environmental indicators are presented in Fig. 5A (comparing OPEX to GW), Fig. 5B (comparing CAPEX to GW), Fig. 5C (comparing OPEX to FRS) and Fig. 5D (comparing CAPEX to FRS).

Based on Fig. 6A, the eco-efficiency analysis demonstrates a distinct relationship between the OPEX and the global warming potential (GW) of the bio-oil produced across different Scenario scenarios and production capacities. Here, the eco-efficiency can be observed where the points fall within the green region, indicating a balance of lower environmental impact per unit of bio-oil produced against the cost incurred.

For S1, the data points shift from the intermediate region into the non-eco-efficient region as production capacity increases from 100 to 600 t/d. This suggests that for S1, while production scales up, operational costs rise without a proportionate reduction in global warming potential, diminishing eco-efficiency. S2 presents an interesting dynamic; the scenario remains consistently in the intermediate region across all production capacities. This indicates a moderate level of eco-efficiency, where neither operational costs nor global warming impacts are optimized to push the scenario into the green zone. S3 show-Scenarios an ideal model at the 300 t/d capacity, aligning perfectly within the eco-efficient zone, denoting a favorable balance between cost and environmental impact. However, at 600 t/d, it regresses slightly into the intermediate zone, showing that although eco-efficiency is still favorable, it is not as optimal as at lower production capacities.

In terms of operational costs alone, it is clear that S3 outperforms the other scenarios at 300 t/d, offering a promising avenue for cost-effective and environmentally sustainable bio-oil production. The analysis reveals that despite increased operational costs with scaling up production, S3 manages to maintain a competitive eco-efficiency profile, particularly at 300 t/d.

Considering the results depicted on Fig. 6B, which illustrates the eco-efficiency comparison for CAPEX versus Global Warming (GW) potential across different Scenario scenarios and production capacities, the most eco-efficient scenarios are S3 at both 100 t/day and 300 t/day production capacities. These scenarios are positioned within the eco-efficient region (green area), indicating a favorable balance of lower GW impact against the capital investment required. In contrast, both S1 and S2 at 300 and 600 t/day, which reside in the non-eco-efficient zone (red area), represent less favorable eco-efficiency due to their higher GW impacts relative to the CAPEX. The remaining scenarios fall into the intermediate zone, suggesting that there is potential for these Scenarios to shift towards greater eco-efficiency with adjustments to either reduce GW impact or CAPEX.

It is noteworthy that for S1 and S2 at 300 t/day, minimal improvements could potentially move these scenarios into the intermediate or even eco-efficient region, as they are on the transition point of the eco-efficient zone. For the 600 t/day scenarios, focusing on reducing the carbon footprint could significantly improve their eco-efficiency standings due to their comparatively higher costs. The findings imply that, especially for S3, higher production capacities tend to enhance eco-efficiency, suggesting an alignment of economic and environmental benefits at larger scales of operation. Conversely, for S2, there seems to be an optimal point for production capacity that balances cost and environmental impact, likely between 100 and 300 t/d.

Fig. 6C shows the eco-efficiency analysis comparing OPEX against fossil resource scarcity (FRS) for various Scenario scenarios and production capacities offers insights into the balance between economic investment and environmental impact in terms of resource depletion. This suggests that, in terms of the balance between financial expenditure and the conservation of fossil resources, this particular scenario operates most optimally.

For S2 and S3 at 300 t/d, there is potential to enhance their positioning within the eco-efficiency framework. This would involve reducing operational costs which, if achieved, could shift these Scenarios further into the eco-efficient region, demonstrating better resource efficiency and lower environmental impact per unit of biofuel produced.

The rest of the scenarios are situated in the intermediate zone, indicating that while they are not the most eco-efficient, they are not significantly straying towards inefficiency either. To transition these scenarios into the eco-efficient domain, a decrease in the fossil resource depletion score would be required. Essentially, this means adopting strategies that further minimize the reliance on fossil-based inputs or improve the yield of biofuel per input unit, thereby lowering the overall FRS metric.

Lastly, Fig. 6D represents the eco-efficiency analysis of CAPEX with fossil resource depletion (FRS) shows that only the scenario of S3 with a production capacity of 300 t/d falls into the eco-efficient category.

For the 100 t/day production capacity, it appears that S3 is positioned within the eco-efficient zone, indicating that it achieves a balance between capital expenditure and conservation of fossil resources. S1 and S2, while showing higher CAPEX, also demonstrate higher FRS, placing them outside of the desired eco-efficient zone.

Looking at the 300 t/day capacity, the trend remains consistent for S3, maintaining its position within the eco-efficient region. This indicates that even with increased production, S3 effectively manages its resource impact while scaling up. However, for S1 and S2 at this production level, there is a visible shift towards increased FRS, which points towards a less favorable eco-efficiency profile, potentially requiring strategies to reduce the fossil resource impact.

At the highest assessed capacity of 600 t/d, all Scenario scenarios show an increase in FRS, although S3 still maintains the closest proximity to the eco-efficient zone. This suggests that S3 process might be more adaptable to scale without excessively increasing its dependency on fossil resources. On the other hand, S1 and S2 at this capacity level move further into the region indicative of lower eco-efficiency, suggesting a diminishing return on investment from an environmental sustainability standpoint. The analysis indicates that as production capacity increases, the challenge of maintaining eco-efficiency becomes more pronounced.

To this end, considering all the eco-efficiency analysis developed, it could be stated that S3 is the most eco-efficient alternative and it becomes evident that leveraging higher production scales, specifically within the 300 to 600 t/d bracket, amplifies eco-efficiency. The amplification is attributable to a synergetic interplay between economic expenditure and reduced environmental footprint, providing a more sustainable production paradigm. Conversely, S1 displays a clear imperative for enhancement in both environmental performance and cost management to reach an eco-efficient condition. A noteworthy observation is the tendency of S1 to manifest improved eco-efficiency metrics at escalated production volumes, signifying that economies of scale play a critical role in bolstering its sustainability profile. Distinctly, S2 diverges from this pattern. Here, the analysis infers that a moderate production capacity, between 100 and 300 t/d, is more favorable to achieving eco-efficiency. This phenomenon may be indicative of an inherent limitation within S2′s process design or operational methodology, which prevents it from obtaining the conventional benefits associated with increased production scales.

4.4. Circularity assessment

With the aim to determine the best technological performance and the most appropriate production capacity, a comparison has been made using the most relevant indicators, as shown in Fig. 7.

Analyzing the performance indicators (Fig. 7A), S2 emerges with the highest impact, indicating it places the greatest strain on the depletion of critical raw materials, contributes most significantly to environmental loads in terms of climate change, and incurs the highest production costs for bio-oil. When examining the production capacities for S2, the lowest level, 100 t/day, yields the poorest results for the “critical raw materials of bio-oil” and “climate change impact of bio-oil” indicators. Meanwhile, the medium level, 300 t/day, is associated with the highest impact on bio-oil production costs. Consequently, among the scenarios evaluated, S2 is the least attractive from a circularity standpoint. In contrast, when seeking the most suitable, sustainable, and profitable option, S3 delivers the best outcomes within the same production capacity and excels at a high production level, specifically 600 t/day. As for S1, it shows the most promise as production capacity increases, suggesting that aiming for medium or high production rates could be most advantageous for achieving better circularity.

Regarding the “resources input & output” indicators (Fig. 7B), the values obtained are similar to those previously discussed, with one notable exception: increasing production capacity leads to improved efficiency in terms of profits generated. On the other hand, focusing on the technology configuration, S3 and S1 provide the best performance, with analogue results, except for the lowest production capacity of 100 t/day. At this level, S1 obtains the best circularity profile.

Finally, regarding the use of energy-related indicators (Fig. 7C), it could be seen that choosing one technology over another does not significantly affect the indicator value. However, S3, with a production capacity of 600 t/d, stands out as the best scenario in the Scenario of the “renewable energy indicator”, given the fact that, as higher the score, as better for circularity, meaning that higher amount of energy is produced by the biooil per kWh of energy required for the process. Conversely, when it comes to “circular process energy intensity”, while the different technological performances do not vary greatly, the choice of

Table 6
Individual and global scores obtained for the composite indicator.

Environmental	S1-100	S1-300	S1-600	S2-100	S2-300	S2-600	**S3-100**	**S3-300**	**S3-600**
CI_{LCA}	0.88	0.88	0.88	0.93	0.93	0.84	0.99	1.00	1.00
Economic	**S1-100**	**S1-300**	**S1-600**	**S2-100**	**S2-300**	**S2-600**	**S3-100**	**S3-300**	**S3-600**
CI_{LCC}	0.06	0.16	0.27	0.03	0.27	0.28	0.61	1.00	0.64
Circularity	**S1-100**	**S1-300**	**S1-600**	**S2-100**	**S2-300**	**S2-600**	**S3-100**	**S3-300**	**S3-600**
CI_{CA}	0.59	0.59	0.68	0.63	0.61	0.72	0.65	0.63	0.89
$\mathbf{CI_{LCA\text{-}LCC\text{-}CA}}$	**0.51**	**0.54**	**0.61**	**0.53**	**0.60**	**0.61**	**0.75**	**0.88**	**0.84**

production capacity has a significant impact. In this Scenario, a higher production capacity yields the most favorable results in terms of energy circularity.

Overall, the comprehensive analysis of performance, resources input & output, flows and energy-related indicators across Fig. 7 highlights the distinct characteristics and circularity performance of the three Scenario scenarios at varying production capacities. S1 and S2 emerges as the least favorable option concerning circularity and sustainability, while, n contrast, S3 demonstrates once again a good circularity performance, particularly at a production capacity of 600 t/d, highlighting its efficiency and circularity in both resource utilization and energy usage. To this end, it could be concluded that this analysis not only underscores the pivotal role of technological choice and production scale in optimizing bio-oil production but also emphasizes the importance of strategic planning, i.e. opting for renewable resources, working on waste reduction strategies, etc., in achieving sustainable and efficient bio-oil production systems.

4.5. Composite indicator

As proposed in the eco-efficiency analysis, it has been thought of combining not only environmental values with economic indicators, but also circularity indicators. In this aspect, both the individual values and the overall value of the indicator have a value between 0 and 1, where 0 is the least sustainable and circular process and 1 is the most sustainable and circular one. The purpose of this analysis is to confirm that the scenario that shows the best results in the different pillars evaluated individually, also provides the best composite indicator score when combined.

Table 6 shows the values obtained for each scenario and each production capacity. As can be seen, for the individual indicator CI_{LCA}, the increase in production capacity does not have a significant effect, with S3 showing the best performance, followed by S2, with the only exception of the production capacity of 600 tons/d, for which, as already observed in the previous results, the score is worse, being S1 the one that obtains a better value for this production capacity.

As far as the individual CI_{LCC} indicator is concerned, the variation between the values is very significant; it is worth considering that for this calculation an analogous MSP has been considered for all the Scenarios studied, i.e. \$5.5/kg, with which all the scenarios show a positive NPV for all the production capacities. The fact that S3 is the most economically viable and that it requires a lower MSP of the bio-oil produced, turns it into the scenario with the highest score of the indicator, much higher compared to Scenarios 1 and 2. On the other hand, among these, it is S2 that has a better result for the productive capacities of 300 and 600 t/d, while S1 for 100 t/d.

Finally, in the aspect of circularity, CI_{CA}, the values obtained are not so different, with a range between 0.59 and 0.89. Again, it is S3 the one that reaches the highest value among all the production capacities evaluated, followed by S2, while S1 shows the lowest degree of circularity.

When evaluating the individual indicators, the same trend is observed when obtaining the final value of the $CI_{LCA\text{-}LCC\text{-}CA}$ composite indicator, with S3 clearly performing better when circularity and sustainability are combined. Although all production capacities maintain S3 as the best technological performance, it is the 300 t/d production capacity that shows the best score. Therefore, given the analysis of the results, it is confirmed and demonstrated that the preference ranking of the most viable scenario to be developed on a large scale is maintained, and therefore the combination of sustainable and circular perspective is in line with the previous individual results.

5. Conclusions

Several models for the valorization of black liquor (BL) to produce biofuel for the aviation and shipping sectors have been evaluated from an environmental, economic and circular point of view. For the environmental assessment, S3 stood out as the technological model with the lowest environmental impact in all the categories studied. On the other hand, the increase in production capacity has a positive effect in S1 and 3, while for S2, the choice of 600 t/d implies a greater impact on the categories. The same trend has been observed when assessing the economic perspective: S3 is the most economically viable and efficient, as it requires a much lower minimum selling price (MSP) of the biofuel compared to the other scenarios. On the other hand, regarding production capacity, the increase in production capacity has a positive effect on the economic viability in S1 and S3, this latter turned out to be the most eco-efficient scenario. With respect to the circularity perspective, it was observed that increased production capacity implies better circularity performance, while for the energy-related indicators, production capacity does not have a significant effect, with the technological model contributing the most. Again, in terms of circularity, S3 is the best performer, with the values obtained for the indicators “renewable energy” and “cost of bio-oil produced” being significantly better compared to S1 and S2. Finally, all individual scores were combined into a composite indicator and S3 showed a higher overall indicator value across all production capacities.

As an overall conclusion, it is considered that this article shows a promising valorization model to be applied in the biofuels value chain, which has not only demonstrated its environmental suitability given its comparison with fossil fuels such as naphtha or kerosene but has also proven to be economically viable production scenarios with a high potential to be categorized as circular models.

CRediT authorship contribution statement

Ana Arias: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis. **Chrysanthi-Elisabeth Nika:** Writing – review & editing, Supervision, Conceptualization. **Gumersindo Feijoo:** Writing – review & editing. **Maria Teresa Moreira:** Writing – review & editing, Supervision, Conceptualization. **Evina Katsou:** Writing – review & editing, Conceptualization.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2024.155335.

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