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Evaluation of environmental and economic hotspots and value creation in multi-product lignocellulosic biorefinery



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ABSTRACT

This paper presents systematic analysis of value creation chains and their economic and environmental hotspots within a multi-product biorefinery with the primary goal to promote sustainable biorefining. Lignocellulosic biorefinery producing ethanol, crude lignin oil (CLO) and electricity was analysed. The methodology involves transformation of technological model into an input-output one with the use of matrix notation for the analysis of economic and environmental attributes along value chains. The results show that the accumulation of biomass through energy supply grows by a factor of 1.2 for ethanol and electricity, and by 1.4 for CLO value chains, of which solid recovery, lignin solvolysis and biomass pretreatment are responsible for the most significant growth indicating the necessity for energy optimization of those steps. The analysis reveals a superfluous role of infrastructure in pretreatment and lignin drying processes. Of infrastructural costs related to the equipment required for the pretreatment step, wastewater treatment (WWT) facility is responsible for 58%, and of the costs of lignin drying, the combined heat and power plant is responsible for 56%. WWT determines 75% of infrastructural GHG emissions attributable to pretreatment, and 57% of those related to lignin drying. This points at an advantage of a biorefinery concept involving the removal of lignin fraction before the valorization of carbohydrates. Another hotspot, the lignin solvolysis technology, shows environmental and economic advantages of its further optimization in terms of production costs and GHG emissions. The proposed method is helpful for analyzing economic and environmental hotspots in new biorefinery concepts and integration pathways.

1. Introduction

The transition to a more sustainable production that allows for both economic prosperity and the protection of natural environment was highlighted by the Sustainable Development Goals formulated by the United Nations and reflected in the 2030 Agenda for Sustainable Development [1]. Although bioeconomy was recognized as a "transformative force" on the way towards sustainable development [2], its own sustainability issues can be also pivotal in this transition [3].

The development of efficient bio-based value chains, with a biorefinery being the core part of those chains, plays a key role in a successful transition to the sustainable and circular bioeconomy. By definition, biorefining refers to "the processing of biomass into a portfolio of marketable bio-based products, which could include coproduction of food and feed, chemicals and materials and bioenergy" [4]. The biorefinery can benefit further from the use of second generation-feedstock, such as wood and agricultural residues, as it could improve economic and environmental profiles of commodities produced [5]. Furthermore, the development of a multi-product biorefinery can bring even more sustainability potential to the biorefinery concept as it boosts resource efficiency via parallel valorization of different side-flows [6].

Yet, the slow progress in the commercialization of lignocellulosic biorefineries involving the biochemical conversion pathway points at the immaturity of existing biorefinery concepts. There were several commercial-scale cellulosic ethanol biorefineries launched since 2013, such as Crescentino, Italy, Maabjerg Energy Concept Consortium, Denmark, Project Liberty (POET-DSM), Abengoa and Dupont biorefineries in the USA, which encountered difficulties in their functioning and, currently, many of them are either idle or on hold [7]. The

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recalcitrant structure of lignocellulosic biomass, the selection of a low-cost efficient pretreatment technology, the overall process complexity and financial issues were identified among the main reasons for the slow biorefinery commercialization [7,8].

The failures experienced by the entities as above, on the other hand, stimulate analysis aimed at the identification and improvement of the most critical, economically or environmentally, biorefinery processes, or hotspots. The life cycle assessment (LCA) methodology using standardized procedures to account for the environmental impact of products is widely applied for the environmental assessment of biorefineries [9], while techno-economic analysis (TEA) converting technology-related data into cost-related information, is commonly applied for the evaluation of economic hotspots [10]. The term "hotspot" can be determined as a point where "major environmental problems or costs" appear on a product's life cycle [11], or as "life cycle stage, process or elementary flow which accounts for a significant proportion of the impact of the functional unit" [12], or as "key influencing factors" [13].

As one of the LCA goals is to improve environmental performance of a product [14], the evaluation of environmental hotspots might be involved in such analysis. Several research efforts focused on environmental hotspots, like the studies on biorefineries producing adipic acid [15], algae-derived biodiesel [16], lignin nanoparticles [17], lactic acid [18], as well as those exploring enzyme production routes for a biorefinery [19]. Corona et al. [20] defined the hotspots of a biorefinery based on the environmental impact related to the process and biorefinery stage; Bello et al. [19] concluded that enzyme production processes should be included in the system boundary as they could be identified as the major hotspot of a biorefinery. Koch et al. [17] evaluated hotspots associated with the use of thermal energy and virgin solvent. Ögmundarson et al. [18] applied the cradle-to-grave LCA for identifying the environmental impact hotspots of lactic acid production systems and reported the large influence of feedstock production and biorefinery processes on the environmental profiles of outputs. It should be noted that, generally, the mentioned studies cover early-stage designs, while keeping aside more rigorous models, including services required to run a stand-alone biorefinery.

TEA often involves the calculation of a minimum selling price for biorefinery products, i.e. product price that balances production costs and biorefinery revenues by using discounted cash flow method [21–24]. Given a certain economic objective function, such as net present value, TEA can be coupled with optimization that allows to conclude on a set of selected biorefinery parameters [25,26].

Although the biorefinery design can benefit from a simultaneous TEA and LCA, in which a more explicit range of indicators may be applied to value creation chains, LCA studies of biorefineries are often disconnected from TEA [27]. As a consequence, these type of studies are less frequently encountered in literature. Among the latter group of studies, there are simultaneous TEA and LCA applied to an energy-driven biorefinery [28], ethanol and cellulosic isobutanol biorefineries [29,30], marine biofuel production [31], integrated mango waste biorefinery [32]. Shi and Guest [27] suggested an integrated modelling framework, allowing to analyse the environmental impacts of different biorefinery designs and with different assumptions, and demonstrated the application of this framework to a sugarcane ethanol biorefinery. The BioSTEAM-LCA open-source package proposed by this study was created in the Python environment, and it originates from the BioSTEAM package applicable to TEA of biorefineries under uncertain conditions [33]. Undoubtedly, open-source software can significantly contribute to the sustainable development of the biorefining industry, though more specific programming skills may be needed for modelling.

A somewhat different approach was demonstrated by Soltanian et al. [34], who determined biorefinery hotspots based on the exergoeconomic analysis to define principal cost-driven losses that affected such a system. The authors systematically apply a wide range of indicators to all biorefinery subunits, including those related to infrastructure, and highlight the crucial role of the steam generation unit in the total cost of the biorefinery.

By definition, a biorefinery involves a series of processing steps aimed at adding value to each intermediate output. Quite often, the main conversion processes are to be complemented with a comprehensive infrastructure serving all energy and ecological needs of a biorefinery due to its typically remote location. As a result, the value creation process may boost costs and environmental burdens added at each processing step, and this requires a uniform assessment of related hotspots.

In the current study, we suggest a systematic approach for the evaluation of hotspots of a stand-alone multi-product biorefinery with a focus on product individual value chains, and involve the matrix notation to this end. Thus, while most of studies focus on biorefinery hotspots with final products in sight, the proposed methodology allows for the value-specific analysis of hotspots, including the evaluation of the influence of the infrastructure on the value chains.

The proposed approach is mainly aimed at complementing the already existing TEA and LCA of a multi-product biorefinery, as, given its uniformity, it can be applied to a variety of indicators that can reveal the most critical processing stages. A lignocellulosic biorefinery simultaneously producing ethanol, crude lignin oil and electricity is selected in this study as a representative case of a multi-product biorefinery for demonstration purposes.

2. Methodology

Starting from a biorefinery concept, the methodological framework includes several subsequent steps as presented in Fig. 1.

The first step of the methodological framework includes the development of a biorefinery inventory that implies the collection of data on the use of chemicals and energy by biorefinery processes, as well as on materials produced by them. After computing allocation factors, those data are further transformed into a biorefinery input-output model and in a coefficient allocation matrix (Fig. 1). At the second step, the allocation matrix obtained is converted into the coefficient matrix of cumulative allocation effects that contains basis for the further evaluation. The next step includes the computation of cumulative attributes associated with individual value-chains and the definition of the related impact indicators. The final step of this methodology involves the interpretation of the results aimed at revealing biorefinery hotspots, if any can be found. The system boundary was defined as a cradle-tobiorefinery gate. The details on the methodology involved are provided in the following sections. The methodological basis of this study was presented at the EUBCE 2021 [35].

2.1. Biorefinery model

This paper considers a lignocellulosic biorefinery that simultaneously produces three co-products: ethanol, crude lignin oil (CLO) and electricity.

The existing cellulosic ethanol biorefineries are currently at the technology readiness level of seven or higher [4]. For analytic purposes, we use a reference cellulosic ethanol biorefinery model developed by the *National Renewable Energy Laboratory, NREL* [23,24]. The biorefinery utilizes agricultural residues like corn stover that includes all non-edible parts of corn gathered above the ground, e.g. leaves, stalks, husks, cobs, etc. [23]. The design includes the biomass pretreatment technology, where first hemicellulose carbohydrates are converted into soluble sugars, and cellulose chain length and crystallinity are reduced [23]. After the pretreatment, the slurry undergoes enzymatic hydrolysis and fermentation, allowing to hydrolyse cellulose, and to convert sugars to ethanol [23]. In the solids recovery area, beer is initially subjected to distillation, where moist lignin cake is separated at the column bottom, while ethanol obtained at the top of the latter is further de-watered and purified.

Fig. 2 presents this biorefinery as an input-output model, separating



Fig. 1. The methodological framework.

the main and infrastructural processes and subdividing these processes to the extent that is needed to achieve the research goal.

Currently, the major proportion of technical lignin obtained in a cellulosic biorefinery is burnt for energy purposes [36], however, this lignin features a high potential for its valorization towards chemicals and fuels, including the utilization of lignin-based oil as a marine bunker fuel that constitutes a promising pathway to upgrade technical lignin [37,38]. The technology for valorization of lignin to CLO was developed by Vertoro, a Dutch company [39] and tested in a multi-purpose pilot plant at the Brightlands Chemelot Campus in 2020 [40]. This technology has been integrated into the reference cellulosic ethanol biorefinery design using the Aspen Plus ® software (Fig. 3).

Based on our previous analysis, it was assumed that about 40% of the wet lignin residue produced is purposed for further valorization, while the rest is sourced to the combined heat and power plant (CHP) for energy recovery [41]. Such ratio allows to supply high and low pressure steam to all biorefinery users and to avoid the imports of any external energy resources. The lignin stream aimed for CLO production is initially dried and then, subjected to mild solvolysis with methanol [39]. The lighter lignin fraction, obtained after solvolysis, which is referred to in this paper as CLO is collected as a product with a lower heating value of about 24 MJ kg⁻¹ and lignin and methanol content of 78 and 10 wt%, respectively, while the insoluble heavier fraction (char) is sent back to the CHP plant.

The considered biorefinery includes an extensive infrastructure that allows to serve all its energy needs, as well as to provide required environmental services (Fig. 2).

The approach to model biorefinery inventory was demonstrated in

our previous paper [42]. The detailed biorefinery inventory breakdown is provided in the supporting information.

2.2. Matrix approach

The matrix approach allowing to track environmental attributes, such as GHG emission, was discussed in more detail in a previous paper [42], and, in the current study, it is extended to include a wide range of economic and environmental attributes, and to make it applicable to the analysis of individual value chains within the biorefinery boundary. The methodological basis of this study was presented at the EUBCE 2021 [35].

The main stages of the methodology proposed are as follows: (i) reckoning the cumulative attributes of intermediate streams and final products, (ii) assigning cumulative attributes to individual value chains, and (iii) analysing the biorefinery hotspots.

2.2.1. Cumulative attributes

Since a biorefinery can be defined as an industrial network of interconnected technological processes, the first methodological step involves the transformation of that technological network into a mathematical input-output model. It should be noted that the depth of such model particularization will be defined by data availability and by the study goal.

The crucial feature of a lignocellulosic biorefinery is that processes may result in multiple products. In the current biorefinery, there are three such processes (Fig. 2): solids recovery, lignin solvolysis, and the CHP plant that subsequently requires applying the appropriate



Fig. 2. Biorefinery model in an input-output form.

allocation procedure for these processes. The latter allows defining the share of economic and environmental attributes to be assigned to certain output.

The allocation method chosen to partition attributes within the main processes is based on the mass of carbohydrates (cellulose, hemicellulose, arabinan, galactan and mannan) and of lignin in the processed corn stover. This method looks very logical to underline the attributional nature of the current LCA study, as process-scaling functions are very much related to the carbohydrate and lignin content. For instance, enzyme loading and the mass of virgin methanol depend on the mass of cellulose and lignin processed, respectively [23,39]; the mass of sulfuric acid relates to the dry mass of the biomass containing mainly structural carbohydrates and lignin. In an earlier study, it was shown that LCA results hardly depend on the allocation methods applied to main conversion processes, unless the total mass and economic allocation is applied [42].

The energy content allocation method [43], which accounts for the useful energy of products, is applied to CHP plant products, as this method seems to be more appropriate for the allocation of both economic and environmental attributes:

$$AF_{i} = \frac{F_{i} \cdot (h_{i} - h_{ref})}{\sum F_{i} \cdot (h_{i} - h_{ref})}$$
(1)

where F_i is the mass flow of ith steam product; h_i and h_{ref} are the specific enthalpy of ith steam product as it is extracted and at the reference conditions, respectively. For obtaining the allocation factor for electricity, the work potential is taken equal to the power produced [43]. Allocation factors applied in this study are listed in Table 1.

The development of a coefficient allocation matrix (matrix *A*) for a biorefinery and its properties were described by Obydenkova et al. [42]. This matrix maps the transfer of attributes from a process input to a

process output flow for the entire biorefinery.

The equation provided by Suh [44] was used to account for direct and indirect inputs and track them throughout the system:

$$\boldsymbol{x} = \boldsymbol{y} + A \cdot \boldsymbol{y} + A^2 \cdot \boldsymbol{y} + A^3 \cdot \boldsymbol{y} + \dots = (I - A)^{-1} \cdot \boldsymbol{y}$$
⁽²⁾

where A is $n \times n$ input-output matrix, x is $n \times 1$ vector of output level, y is $n \times 1$ vector of final deliveries, I is the $n \times n$ identity matrix.

Equation (2) can be used in the same way to track attributes along the biorefinery value chains, as the effect of the transfer of attributes from one flow to another is defined via the multiplication of transfer coefficients defined in matrix A:

$$\boldsymbol{e}_{\boldsymbol{c}} = \boldsymbol{e} + A \cdot \boldsymbol{e} + A^{2} \cdot \boldsymbol{e} + A^{3} \cdot \boldsymbol{e} + \dots = (I - A)^{-1} \cdot \boldsymbol{e} = C \cdot \boldsymbol{e}$$
(3)

where:

- $e_c = (e_{c1}, e_{c2}, ..., e_{cn}) \in \mathbb{R}^n$ is the vector representing cumulative attribute (economic or environmental) related to the streams within a biorefinery boundary;
- $e = (e_1, e_2, ..., e_n) \in \mathbb{R}^n$ is the vector of input attribute (economic or environmental);
- A is the non-negative coefficient allocation matrix;

• $C = (I - A)^{-1}$.

The matrix $(I - A)^{-1}$ in eq. (2) and eq. (3) is the Leontief inverse coefficient matrix. Regarding the biorefinery under study, this matrix reflects the share of cumulative attributes to be assigned to each flow within the system boundary, and, similarly to previous studies, in the current paper it is referred to as the coefficient matrix of cumulative allocation effects.

The relation between the Leontief inverse matrix in the input-output analysis and the inverse of technology matrix in LCA was thoroughly



Fig. 3. The integration of lignin drying and solvolysis technology into the cellulosic ethanol biorefinery (BRF – biorefinery, LRm – moist lignin residue, LPS – low pressure steam, HPS – high pressure steam, CW – cooling water, HEX – heat exchanger).

Table 1

Process	Product	Stream ID	Allocation factor
Solids recovery	Diluted ethanol (AF _{Ethanol})	DE	0.789
	Moist lignin residue (AF _{Lignin})	LRm	0.211
Lignin solvolysis	Crude lignin oil (AF _{CLO})	CLO	0.595
	Char (AF _{Char})	Char	0.405
CHP plant	Electricity (AF _{Electricity})	EL	0.309
	High-pressure steam to pretreatment (AF _{HPS1})	HPS1	0.201
	High-pressure steam to solvolysis (AF _{HPS2})	HPS2	0.008
	Low-pressure steam (AFLPS)	LPS	0.481

considered by Heijungs and Suh [45].

A document demonstrating the development of the coefficient allocation matrix is provided in the supporting information.

2.2.2. Individual value chains

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As it can be seen from eq. (3), vector e_c shows the cumulative amount of attribute assigned to each stream within biorefinery boundary. However, for a sustainable biorefinery design, there is a need to analyse how economic and environmental attributes are being accumulated inside individual value chains. The latter can be obtained via the multiplication of diagonal allocation matrix $diag(y_k)$, specific to a value chain, by the vector of cumulative attribute:

$$\mathbf{v}_k = diag(\mathbf{y}_k) \cdot \mathbf{e}_c \tag{4}$$

:

where v_k is the vector of a cumulative attribute allocated to the k^{th} product value-chain; y_k is the k^{th} product value-chain allocation vector. In this vector, each element is a product of the downstream non-zero allocation factors and split factors (if any), related to specific value chain. The element of this vector related to the final product will be equal to one, and elements, which do not belong to a value chain will be equal zero (Fig. 4).

2.3. Analysis of economic and environmental hotspots

Following the definitions given by Refs. [11–13], under the term "hotspot" we imply a biorefinery process that either has a considerable impact on the economic or environmental profile of products, or its performance exceeds some reasonable economic or environmental thresholds, and improvement of those processes will have a positive effect on as many biorefinery value chains as possible.

In this study, we suggest several economic and environmental attributes that can be uniformly involved in the systematic analysis of biorefinery hotspots. These attributes are aimed at (i) the evaluation of monetary and environmental profiles of value chains, (ii) the analysis of the role of the feedstock, and (iii) the analysis of the role of the infrastructure required to support the main processes in those value chains. These attributes are explained in more detail below.

A chain value growth of an attribute at any process stage in an individual biorefinery value chain (G, %) can be obtained using the next equation:



SPL - Split factor, from Inventory table: SPL_{LR-SOLV}=0.379 (LR to Solvolysis); SPL_{LR-CHP}=0.621 (LR to C SPL_{Electricity}=0.203 (Electricity for export)

Fig. 4. Development of individual value chain allocation vectors.

$$G = \frac{v_{k(i)} - v_{k(i-1)}}{v_{kfinal} - v_{k0}} \cdot 100\%$$
(5)

where $v_{k(i)}$ and $v_{k(i-1)}$ are cumulative attributes of a stream right before and after a certain process, and v_{kfinal} and v_{k0} are cumulative attributes of the stream at the beginning and the end of a kth value chain. Thus, in eq. (5), the numerator shows an increase of an attribute in a process stage, and the denominator represents the same attribute increase in the entire value chain.

Since the threshold for selected indicators is not always available in the literature, the ranking of those indicators can be considered as a more appropriate approach to this analysis. A rank is assigned to each indicator within all three value chains. The rank can vary from one, which is the most critical case, to four, the least critical one. Finally, ranking results are summarised for all value chains based on the frequency of appearance of main biorefinery processes in the set of the most critical ones, then of the secondary critical ones, and so on.

All results are normalized to a unit of a certain product.

2.3.1. Production cost and GHG emissions through an individual value chain

Production costs and environmental impacts associated with a biorefinery product can be considered as the main metrics of its sustainability. This concept can be applied to all production stages through the value chain of each biorefinery product.

The production cost after each biorefinery process (PC_{ik}) is defined as:

$$PC_{ik} = \frac{E_i + OPEX_i}{P_k}$$
(6)

where PC_{ik} is production cost after the ith biorefinery process; E_i is the annualized capital cost of the ith process; $OPEX_i$ are the operational expenses related to the ith process; and P_k is the amount of final product related to the kth value chain. Thus, the chain value growth of the production cost (G_{PC}) can be found using equation (5).

Annualized capital cost *E* can be found using the next equation:

$$E = \frac{DR \times (1 + DR)^n}{(1 + DR)^n - 1} \times TCI$$
(7)

where DR is the discount rate; n is the project lifetime and TCI is the total capital investment. The latter is defined as the sum of fixed capital investment, including individual installed cost of equipment, the cost of warehouse, site development and piping, project contingency, field expenses, and the other expenses, such as working capital and the cost of land.

As the main concern of the transition towards biomass sourced carbon is the reduction of GHG emissions, the latter has been selected to evaluate the environmental performance of biorefinery products. Among others, this metric was used as the main environmental parameter in different LCA studies of biorefineries [15,46,47], however, any other impact categories can be analysed if needed. The chain value growth of the GHG emissions (G_{GHG}) can be found using equation (5).

Table 2 summarizes the main inputs related to the calculation of production cost and GHG emissions of the biorefinery.

2.3.2. Accumulation of impacts related to biomass

Within the biorefinery under review, biomass is the primary source of bio-molecules to be converted to value-added products, but also it is the source of energy required by main conversion processes and supplementary facilities. The importance of the feedstock intensity analysis for bio-based production routes was highlighted in Ref. [50], noticing that "this metric informs the economic operator how efficient the employed process is in terms of resource utilization capability".

In the current biorefinery design, steam and electricity are being produced from residual streams, such as lignin cake and organic matter contained in the wastewater. The latter undergoes, first, anaerobic digestion, where organic substances are converted to biogas, and, second, aerobic digestion, producing wastewater sludge [23]. Both these streams are sourced to the CHP plant. Concerning primary energy balance, it was found that the biorefinery can yield 43% of lignin residue purposed for a value-added application that implies a remarkable role of biogas in the biorefinery energy supply chain [41].

Ultimately, because energy is supplied from biomass-sourced residuals, the accumulation of biomass allocated along the value creation chains expands steadily, dictating also an inevitable increase in costs and boosting environmental impact associated with feedstock in these value chains. The chain value growth of the impacts related to biomass ($G_{Biomass}$) can be found using equation (5).

2.3.3. The cost and GHG emissions associated with the main and infrastructural processes

While analysing biorefinery hotspots, it seems rational to separate costs and environmental impacts associated with the main and infrastructural processes, and to detect which particular main processes are more responsible for the use of infrastructure. For that, we refer to the concept allowing to define a portion of equipment as settled inside battery limits (ISBL), or that encountered outside battery limits (OSBL).

While ISBL includes facilities where chemical reactions or refining occur, OSBL comprises processes purposed only to support main production activities, i.e. the latter refers more often to infrastructural processes [51]. Following this concept, biomass pretreatment, enzymatic hydrolysis, enzyme production, fermentation, lignin drying, and solvolysis processes are regarded as ISBL, while wastewater treatment facility, CHP plant, colling services, and storage refer to OSBL.

Although the role of infrastructure in the production process can be significant, it should not dominate the latter. Moreover, since ISBL and OSBL processes may feature both costs and environmental impacts, this analysis can be universally used for the evaluation of both economic and environmental hotspots of a biorefinery. Indicators used for this analysis are as follows:

$$CR_{i}(\%) = \frac{Cost \ of \ installed \ equipment \ OSBL_{i}}{Cost \ of \ installed \ equipment \ ISBL_{i}} \cdot 100$$
(8)

Table 2

Economic and environmental parameters of biorefinery.

Parameter, process	Attribute	Unit	Value	Reference
Corn stover	Annual OPEX	M\$	62.1	(a)
	Annual direct	kt	96.6	(b)
	GHG emission			
Feedstock handling and	(CO _{2-eq})	M\$	75.0	(c)
pretreatment	Installed cost	M\$	37.9	(c)
F	Annual OPEX	M\$	10.9	(c)
	Annual direct	kt	11.5	(b)
	GHG emission			
En averatio huduolusio	(CO _{2-eq})	M¢	110	(c)
fermentation, enzyme	Installed cost	M\$	56.9	(c)
production	Annual OPEX	M\$	14.4	(c)
•	Annual direct	kt	14.7	(b)
	GHG emission			
T	(CO _{2-eq})	3.50	07.6	(c)
Lighin residue recovery	I CI Installed cost	IVI\$ M\$	27.0 13.0	(c)
	Annual OPEX	M\$	1.85	(c)
	Annual direct	kt	0	_
	GHG emission			
	(CO _{2-eq})			
Ethanol recovery	TCI	M\$ M¢	23.2	(c) (c)
		IVI.Þ M.S	11./	(c)
	Annual direct	kt	0	_
	GHG emission			
	(CO _{2-eq})			
Ethanol storage	TCI	M\$	4.32	(c)
	Installed cost	M\$ M¢	2.56	(c) (c)
	Annual direct	kt	0.0709	_
	GHG emission		0	
	(CO _{2-eq})			
Lignin residue drying	TCI	M\$	1.00	(d)
	Installed cost	M\$ M¢	0.502	(d)
	Annual OPEX	lvi.ə kt	0.048	_
	GHG emission		0	
	(CO _{2-eq})			
Lignin solvolysis	TCI	M\$	15.4	(d)
	Installed cost	M\$ M¢	7.79	(d)
	Annual OPEX	lvi.ə kt	2.33 5.97	(b)
	GHG emission		0.57	
	(CO _{2-eq})			
Crude lignin oil storage	TCI	M\$	1.45	(d)
	Installed cost	M\$	0.731	(d)
	Annual OPEX	M\$ kt	0.032	_
	GHG emission	ĸ	0	-
	(CO _{2-eq})			
WWT	TCI	M\$	95.7	(c)
	Installed cost	M\$	56.7	(c) (c)
	Annual OPEX	M\$ let	4.43	(b)
	GHG emission	KL	20.5	
	(CO _{2-eq})			
CHP plant	TCI	M\$	119	(c)
	Installed cost	M\$	70.7	(c) (c)
	Annual OPEX	M\$	5.40	(b)
	GHG emission	KL	13.1	
	(CO _{2-eq})			
Process water supply	TCI	M\$	3.50	(c)
	Installed cost	M\$	2.08	(c) (c)
	Annual OPEX	M\$ kt	0.629	(9)
	GHG emission	KL	U	-
	(CO _{2-eq})			
Cooling service	TCI	M\$	5.62	(c)
	Installed cost	M\$	3.33	(c)
	Annual OPEX	M\$	0.302	(C)
		KL	U	-

(continued on next page)

Table 2 (continued)

Parameter, process	Attribute	Unit	Value	Reference
Chilling service	Annual direct GHG emission (CO _{2-eq}) TCI	M\$	3.68	(c)
	Installed cost Annual OPEX Annual direct GHG emission (CO _{2-en})	M\$ M\$ kt	2.18 0.233 0	(c) (c) -
Number of working hours	_	h year ⁻¹	7880	(c)
Project year	-	-	2018	-
Discount rate (DR)	-	-	0.1	(c)
Project lifetime (n)	-	years	30	(c)

^a Based on [24].

 $^{\rm b}$ Calculated using SimaPro® (v.8.0.2) software, based on input data from Ref. [23].

^c Based on [23], converted to the project year, and scaled-down where it is needed (e.g. for CHP plant) based on size factoring exponents method.

^d Calculated using equation method and equipment cost estimate from Refs. [48,49], and based on simulation data from Aspen Plus® (v.10) software. The breakdown of installed equipment cost for lignin drying, solvolysis and CLO storage is provided in the supporting information.

$$OR_i(\%) = \frac{OPEX_{OSBL_i}}{OPEX_{ISBL_i}} \cdot 100 \tag{9}$$

$$ER_i(\%) = \frac{GHG_{OSBL_i}}{GHG_{ISBL_i}} 100 \tag{10}$$

where CR, OR and ER are OSBL to ISBL installed equipment cost ratio, operational expenses ratio and GHG emission ratio, respectively, and the subscript *i* refers to the *i*th process of the biorefinery.

Input data for the analysis of main processes and infrastructure roles are provided in Table 2.

According to Kinney and Gauche [51], the factor range of OSBL for a stand-alone new greenfield plant may account for up to 60–120% of ISBL process costs. This cost range, hence, can be regarded as adequate. However, there is no evidence of suitability of this range for the environmental analysis or operational expenses.

2.4. Tools and methods

The SimaPro® (v.8.0.2) software was used to access the background process-related life-cycle inventory (LCI) databases. Most of the data were obtained from the U.S. LCI database, including data for corn stover, quicklime, diammonium phosphate, sodium hydroxide, corn steep liquor, ammonia, sulfuric acid, and methanol processes. Ecoinvent 3 and Agri-footprint databases were used for glucose and sulfur dioxide processes. The IPCC 2013 v.1.03 impact assessment method with a 100-year time horizon was used for the quantification of GHG emissions. The R



ETHANOL VALUE CHAIN

Fig. 5. Ethanol value chain results.

software [52] was used to convert the coefficient allocation matrix into the coefficient matrix of cumulative allocation effects. The Aspen Plus® software was used to integrate the lignin conversion technology into the reference model and to access technology-related data.

3. Results

3.1. Ethanol value chain

3.1.1. Chain value growth of the production cost (G_{PC})

Starting at the biomass cost of 279 \$ t⁻¹ of ethanol, downstream valorization inflates that cost by a factor of 2.8, resulting in a total of 792 \$ t⁻¹ of ethanol (Fig. 5). The biomass pretreatment process is responsible for nearly 39% of chain value growth (G_{PC}), which is followed by enzymatic hydrolysis, enzyme production and fermentation processes (G_{PC} = 30%), solids recovery (G_{PC} = 22%) and ethanol recovery (G_{PC} = 9%). The operational expenses prevail in the production cost, however, with the dominating share of biomass cost in the pretreatment hydrolysate of 62%, this share will constantly decrease to 41% in the final product.

3.1.2. Chain value growth of the GHG emissions (G_{GHG})

GHG emissions (CO_{2-eq}) associated with the ethanol value chain starts at 16.1 and ends up at 30.4 kg GJ⁻¹, with the most prominent growth provided by the biomass pretreatment technology, of 45% (G_{GHG}), that is followed by enzymatic hydrolysis, enzyme production, and fermentation (G_{GHG} = 26%), solid recovery (G_{GHG} = 23%) and ethanol recovery (G_{GHG} = 6%) (Fig. 5).

3.1.3. OSBL to ISBL installed equipment cost ratio (CR)

The relative analysis of costs brought in by main and supplementary processes shows a superfluous role of infrastructure in the pretreatment technology with the OSBL to ISBL cost ratio of 149%, which exceeds reasonable limits set for a stand-alone new plant (Fig. 5). Of infrastructural costs related to the equipment required to support biomass pretreatment technology, the WWT facility is responsible for 58%, and the CHP plant for 38%. That material figure related to the WWT facility stems from the amount of processed water required to make solid biomass pumpable.

3.1.4. OSBL to ISBL GHG emission ratio (ER)

Similarly to the equipment cost, the pretreatment technology is found crucial in terms of GHG emissions released by infrastructure involved, with the OSBL to ISBL GHG emissions ratio of 137% (Fig. 5), and with an even more pronounced contribution of WWT facility, at 75%. It should be noted that when it comes to the GHG emissions associated with infrastructure, the WWT dominates at all stages of ethanol value chain.

3.1.5. OSBL to ISBL operational expenses ratio (OR)

The biomass pretreatment technology leads in this category, with OSBL to ISBL operational expenses ratio of 43%, mainly due to the contribution of the WWT facility (55%) and the CHP plant (36%).

3.1.6. Chain value growth of impacts related to biomass (G_{Biomass})

The accumulation of biomass can be tracked throughout the whole ethanol value chain and, in total, it grows by a factor of 1.2, of which both pretreatment technology and solid recovery are responsible for the most significant expansion growth (G_v) of 36% and 44%, respectively (Fig. 6). This accumulation points at the energy intensity of said processes.

3.2. Lignin value chain

3.2.1. Chain value growth of the production cost (G_{PC})

Starting at the biomass cost of 107 \$ t⁻¹ of lignin oil, downstream valorization increases that cost by a factor of 4.7, resulting in a total of 502 \$ t⁻¹ of lignin oil (Fig. 7). On the value chain breakdown, the lignin solvolysis process is responsible for nearly 41% of the cost increase, which is followed by the biomass pretreatment process ($G_{PC} = 19\%$), enzymatic hydrolysis, enzyme production and fermentation processes ($G_{PC} = 15\%$), lignin cake drying ($G_{PC} = 13\%$), and solids recovery ($G_{PC} = 11\%$). Operating expenses, including the cost of feedstock, dominate through the lignin value chain. Given the dominating share of biomass cost in the pretreatment hydrolysate of 62%, the contribution of this cost will decrease to 29% in the final product.

The cost of lignin solvolysis technology is nearly equally determined by both the cost of methanol and the installed cost of equipment, where the solvolysis reactor, filtration unit and evaporator are responsible for 39%, 22% and 14% of the total installed cost of solvolysis process, respectively.

As the matrix notation used in this paper allows finding attributes for any stream within biorefinery boundary, it might be also worth finding production costs of such intermediate as lignin cake, as it may define whether downstream valorization is feasible, or not. Given this approach, the production cost of moist lignin residue amounts to 119 \$ t^{-1} , which is within the range of price estimates [41,53], however, it almost doubles for dry lignin residue, to 216 \$ t^{-1} .

3.2.2. Chain value growth of the GHG emissions (G_{GHG})

GHG emissions (CO_{2-eq}) associated with the lignin-based value chain starts at 6.9, and ends up at 22.3 kg $\rm GJ^{-1}$ of crude lignin oil, with the



Fig. 6. Accumulation of impacts related to biomass.



LIGNIN BASED VALUE CHAIN

Fig. 7. Lignin value chain results.

most prominent growth provided by the lignin solvolysis process, of 53%, that is followed by pretreatment technology ($G_{GHG} = 18\%$), enzymatic hydrolysis, enzyme production and fermentation ($G_{GHG} = 10\%$), solids recovery, and cake drying ($G_{GHG} = 9\%$ each), as shown in Fig. 7.

The GHG emission profile of lignin cake in the biorefinery under assessment amounts to 15.6 and 17.4 kg GJ^{-1} of moist and dry residue, respectively, or 128 and 218 kg t⁻¹.

3.2.3. OSBL to ISBL installed equipment cost ratio (CR)

Similar to the ethanol value chain, this analysis highlights the superfluous role of infrastructure not only in the biomass pretreatment but also in the lignin cake drying processes (see Fig. 7). The OSBL to ISBL installed equipment cost ratio amounts to 149 and 123% in the pre-treatment and drying processes, and, while the pretreatment is more responsible for the use of WWT facility that contributes up to 58% of all costs of infrastructure involved, the drying process is more affiliated with the CHP plant that contributes up to 56% of all costs of the required infrastructure. The WWT and CHP related impacts stem from the amount of processed water required to make the incoming solid biomass pumpable, as well as from the necessity to evaporate a significant amount of water downstream from moist lignin residue, which finally can raise questions about the sustainability of such a biorefinery concept.

3.2.4. OSBL to ISBL GHG emissions ratio (ER)

Both biomass pretreatment and lignin drying processes were found crucial in terms of the usage of highly GHG-intensive supplementary facilities featuring the OSBL to ISBL GHG emission ratio equal to 137 and 122%, respectively, and the WWT facility responsible for nearly 75% and 57% of infrastructure involved in the said processes (see Fig. 7).

3.2.5. OSBL to ISBL operational expenses ratio (OR)

Concerning the OSBL to ISBL operational expenses ratio, the results are generally compatible with the ethanol value chain, indicating that the biomass pretreatment technology is the most critical stage of these value chains.

3.2.6. Chain value growth of impacts related to biomass (G_{Biomass})

The accumulation of biomass can be tracked throughout the whole lignin-based value chain and, in total, it increases by a factor of 1.4, with the main contributions shared by lignin solvolysis ($G_v = 37\%$), cake drying ($G_v = 22\%$), and solids recovery process ($G_v = 20\%$) (see Fig. 6). This accumulation is primarily due to the use of carbon-containing residues for energy purposes, that subsequently highlights the energy intensity of said processes.

3.3. Electricity value chain

3.3.1. Chain value growth of the production cost (G_{PC})

Starting at the biomass cost of 0.012 \$ kWh⁻¹, downstream valorization boosts that cost by a factor of 5.3, resulting in a total of 0.063 \$ kWh⁻¹ of electricity produced (Fig. 8). The power generation process is the main contributor to the increase in production costs ($G_{PC} = 61\%$). Biomass pretreatment and enzymatic hydrolysis, enzyme production and fermentation processes amount to a further 16% and 13% of the total cost growth (G_{PC}), respectively, while the solids recovery process brings another 9% into that increase. Electr

LRm

43%

b) GHG emission (CO_{2-eq}), kg/GJ

9.3

16% 31% 16.7



ELECTRICITY VALUE CHAIN

Electr

LRm

63%

\$0.063



3.3.2. Chain value growth of the GHG emissions (G_{GHG})

GHG emissions (CO_{2-eq}) associated with power generation increases by a factor of 3, starting with 5.1 and ending up with 16.7 kg GJ⁻¹ of electricity (Fig. 8), with the most prominent growth of $G_{GHG} = 64\%$ provided by the CHP plant. Pretreatment technology, enzymatic hydrolysis, enzyme production and fermentation and solids recovery are responsible for 17, 10, and 9% of the production cost growth (G_{GHG}).

a) Production cost (PC), \$/kWh

\$0.031

10%

18%

41%

3.3.3. OSBL to ISBL installed equipment cost ratio (CR), GHG emissions ratio (ER), and operational expenses ratio (OR)

The relative analysis of costs and environmental impacts imported into the value chain by both the main and infrastructural processes reveals a superfluous role of infrastructure in the biomass pretreatment process and the CHP plant (Fig. 8). OSBL to ISBL installed equipment cost ratio amounted to 149 and 433% in these two processes, OSBL to ISBL GHG emissions ratio is equal to 137 and 294%, and operational expenses ratio equals 43 and 128%. While the domination of infrastructure related to the CHP plant is logical, the impact of pretreatment technology still exceeds the reasonable limits observed in the literature.

3.3.4. Chain value growth of impacts related to biomass (G_{Biomass})

The accumulation of biomass can be tracked throughout the entire electricity value chain and, overall, it increases by a factor of 1.2, largely due to the contribution of the power generation process itself ($G_v = 35\%$), solids recovery ($G_v = 31\%$) and biomass pretreatment ($G_v = 26\%$) processes (see Fig. 6).

4. Discussion

Table 3 summarizes the results obtained for the three value chains, while Fig. 9 shows the frequency of appearance of main biorefinery processes in the set of the most critical ones, secondary critical ones, and so on.

Basically, the results obtained support existing observations, including those related to a significant impact of biomass on the production costs and GHG emission of biorefinery products, but they also are shedding more light on the relationship between the main and infrastructural processes within a biorefinery, and, thus, can be used as a basis for a further improvement of the biorefinery concept.

As can be seen in Fig. 9, three processes persist among the most critical, with the pretreatment technology leading this category. Although the remarkable role of the pretreatment technology in the total capital investment of a cellulosic biorefinery was highlighted by Cheng et al. [54], the current analysis also uncovers the significant impact of this technology on production costs and GHG emissions of the ethanol value chain, and the crucial role of infrastructure required to support the

Table 3

Results for the ethanol, lignin, and electricity value chains.

Value chain	Chain value growth			The impact of infrastructure			
	Production cost, G _{PC} , %	GHG emissions, G _{GHG} ,%	Biomass-related impacts, G _{Biomass} , %	Installed equipment cost ratio, CR, %	Operational expenses ratio, OR, %	GHG emission ratio, ER, %	
Ethanol value chain							
Pretreatment hydrolysate	39%	45%	36%	149%	43%	137%	
Beer	30%	26%	11%	85%	27%	82%	
Diluted ethanol	22%	23%	44%	104%	34%	105%	
Ethanol	9%	6%	10%	102%	35%	111%	
Lignin value chain							
Pretreatment hydrolysate	19%	18%	16%	149%	43%	137%	
Beer	15%	10%	5%	85%	27%	82%	
Moist lignin residue	11%	9%	20%	104%	34%	105%	
Dry lignin residue	13%	9%	22%	123%	34%	122%	
Crude lignin oil	41%	53%	37%	110%	29%	56%	
Electricity value chain							
Pretreatment hydrolysate	16%	17%	26%	149%	43%	137%	
Beer	13%	10%	8%	85%	27%	82%	
Moist lignin residue	9%	9%	31%	104%	34%	105%	
Electricity	61%	64%	35%	433%	128%	294%	



PR - Biomass pretreatment; EHF - Hydrolysis, enzyme production and fermentation; SR - Solids recovery; ER - Ethanol recover LDR - Lignin cake drying; LSOLV - Lignin solvolysis;



pretreatment process (see Table 3). The WWT facility dominates both costs and emissions associated with services required for the biomass pretreatment stage, which also supports the observation by Tobin et al. [55], reporting a considerable contribution of the digester and the supplement treatment to the total capital costs of the lignocellulosic biorefinery.

The results obtained herein demonstrate that the environmental and economic performance of the biorefinery studied will mainly benefit from an improvement in the pretreatment technology, as this will positively affect the most critical stages in all three value chains.

The second process that very frequently emerges among the most critical ones, is lignin solvolysis. Although the GHG emission profile of CLO obtained (22.3 kgCO_{2-eq} GJ⁻¹) allows decreasing GHG emissions by 76% compared to fossil fuel and, hence, to comply with the threshold of 65% set by the European Renewable Energy Directive [56], this profile can be further improved by the reduction of the highly GHG-intensive methanol content in CLO product. However, considering the expected trade-off between virgin solvent content and thermal energy needed for solvent recycling [17], future biorefinery designs should evaluate thoroughly this trade-off with regard to both economic and environmental performance of the biorefinery.

The solid recovery, including beer distillation and lignin filtration processes, can be regarded as another hotspot of the biorefinery (see Fig. 9), as it showed the worst performance in terms of accumulation of impact related to biomass in the ethanol value chain (Table 3), It was

also found unsatisfactory in terms of the accumulation of impacts related to biomass, and the cost of infrastructure involved. This finding is also in agreement with Cheng et al. [54] who noted a significant role of the lignin separation process in the fixed capital investment of a cellulosic biorefinery.

The drying of lignin residue can be also regarded as a biorefinery hotspot (Fig. 9), as, although it occurred in the second critical rank category, it showed unsatisfactory results in the accumulation of impacts related to biomass, as well as in all indicators related to the economic and environmental performance of the infrastructure (Table 3). As follows from the results, the installed cost of the CHP plant amounts to 56% of the total services involved in the lignin drying process, and both the WWT and CHP facilities play a notable role in GHG emissions associated with the drying step (Fig. 7).

We can also mention enzymatic hydrolysis, along with enzymes production, and fermentation, as well as ethanol recovery as unsatisfactory in both economic and environmental sense. While hydrolysis influenced mainly production cost and GHG emission of the ethanol value chain (Table 3), ethanol recovery bunch of processes was the second in terms of operational expenses and GHG emission associated with infrastructure, where the WWT facility took the leading role (at 63%) in the environmental impact associated with the recovery process (Fig. 5).

The following recommendation can be drawn based on the hotspot analysis of the lignocellulosic multi-product biorefinery. The environmental and economic hotspots associated with the biomass pretreatment, solids recovery and lignin cake drying processes stem from the amount of water circulating in the biorefinery. That water is mainly needed to ensure the pumpability of solid biomass material, however, it subsequently defines the energy intensity of such downstream processing stages as distillation, evaporation and lignin cake drying. As the main role of the pretreatment technology is to decompose the rigid structure of biomass liberating lignin and making hemicellulose and cellulose available for the downstream hydrolysis, the removal of lignin before the hydrolysis step would not only decrease the amount of water needed for the valorization of those carbohydrates, but also would allow improving the product recovery, as well as the lignin drying steps.

5. Limitations

Along with the benefits provided by the presented approach to analyse the biorefinery hotspots, there are also limitations imposed by this method. The main limitation refers to the allocation procedure.

Although the application of the same allocation method to both economic and environmental parameters makes this analysis more uniform, the choice of the method itself can be considered somewhat arbitrary. For instance, while the Greenhouse Gas Protocol recommends using the allocation method based on the fuel efficiency for heat and electricity production, and energy content or the working potential method for the sensitivity analysis [43], four methods are suggested for cost allocation in CHP plants [57]. Among them are the residual value method, electrical equivalent, exergy and calorific methods, and the selection of an appropriate allocation methodology would be defined "case-by-case" [57]. However, for instance, the recent LCA study highlighted the importance of the choice of allocation methods applied to the CHP plant [42]. Overall, the allocation procedure was identified as the main source of uncertainty in the attributional LCA [58] and, given the prevailing multifunctionality, the bioeconomy sector would require the development of consistent allocation procedures [59].

Another limitation refers to the selection of thresholds for the analysis of economic and environmental indicators. For instance, while there is an understanding of a reasonable range of installed equipment costs referred to the OSBL of a stand-alone new greenfield plant [51], the reasonable range of environmental impacts brought into the value chains by the infrastructure is unavailable. More studies are needed in order to depict this realistic range of impacts associated with infrastructure.

6. Conclusions

This paper evaluates environmental and economic hotspots of a multi-product lignocellulosic biorefinery with product portfolio including biofuels and electricity. To this end, this study proposes a novel matrix-based approach for the analysis of value creation in such biorefinery and defines a set of relevant environmental and economic indicators to analyse the role of feedstock, as well as the role of main biorefinery processes and infrastructure in those value chains, and their monetary and environmental profiles.

Within this analysis, a significant impact of biomass on production cost and on greenhouse gas (GHG) emission of outputs is found, which is in line with previous studies. However, the named impact is more prominent for ethanol rather than for crude lignin oil and electricity, whose value chains are also affected by other biorefinery processes. Solids recovery, lignin solvolysis and biomass pretreatment are among the main processes responsible for the accumulation of biomass, largely through the biorefinery energy supply network that points at the necessity for energy optimization of said processes.

This study reveals the exorbitant role of infrastructure required to support the biomass pretreatment process, which is the main identified biorefinery hotspot. This redundancy is attributable mainly to the wastewater treatment facility dominating both costs and emissions associated with services required to run the process. Hotspots associated with the pretreatment stage are based on the large amount of water circulating in the biorefinery that is mainly needed for making flows pumpable and ensure proper solid loading in enzymatic hydrolysis. A decrease in water consumption can be achieved via the removal of lignin from biomass before the hydrolysis step. This improvement, in fact, will have a double positive effect, as it will also reduce energy used by the recovery of solids, the other biorefinery hotspot.

Concerning hotspots associated with the lignin value chain, the lignin solvolysis technology would need a further optimization in both production costs and GHG emission. Although the GHG emission profile of lignin oil allows decreasing the GHG emissions by 76% compared to fossil fuel, it is concluded that a significant environmental improvement can be attained in the lignin value chain via a reduction of methanol content in the lignin oil. However, trade-offs between energy use and solvent recycling need to be analysed along with the analysis of economic and environmental profiles of that value chain.

The results demonstrate the effectiveness of the matrix notation for the analysis of value creation and of hotspots within a biorefinery. The proposed method can be applied to the analysis and optimization of new biorefinery concepts and new integration pathways. Among limiting factors, we stress the expected sensitivity of results to allocation procedures that should be properly addressed during analysis.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biombioe.2022.106394.

References

- UN, The sustainable development Agenda. https://www.un.org/sustainabledevelopment/development-agenda/.
- [2] IACGB, Expanding the Sustainable Bioeconomy Vision and Way Forward. Communiqué of the Global Bioeconomy Summit 2020, 2020. https://gbs2020. net/wp-content/uploads/2020/11/GBS2020_IACGB-Communique.pdf.
- [3] T. Heimann, Bioeconomy and SDGs: does the bioeconomy support the achievement of the SDGs? Earth's Future 7 (1) (2019) 43–57.
- [4] EC, EU Biorefinery Outlook to 2030 : Studies on Support to Research and Innovation Policy in the Area of Bio-Based Products and Services, 2021.
- [5] IEA Bioenergy, Biorefineries: Adding Value to the Sustainable Utilisation of Biomass, IEA Bioenergy, 2009.
- [6] EC, A Sustainable Bioeconomy for Europe : Strengthening the Connection between Economy, Society and the Environment, updated bioeconomy strategy, 2018.
- [7] T. Raj, K. Chandrasekhar, A. Naresh Kumar, J. Rajesh Banu, J.J. Yoon, S. Kant Bhatia, Y.H. Yang, S. Varjani, S.H. Kim, Recent advances in commercial biorefineries for lignocellulosic ethanol production: current status, challenges and future perspectives, Bioresour. Technol. 344 (Pt B) (2022) 126292.
- [8] Z. Usmani, M. Sharma, A.K. Awasthi, T. Lukk, M.G. Tuohy, L. Gong, P. Nguyen-Tri, A.D. Goddard, R.M. Bill, S.C. Nayak, V.K. Gupta, Lignocellulosic biorefineries: the current state of challenges and strategies for efficient commercialization, Renew. Sustain. Energy Rev. 148 (2021).
- [9] E. Gnansounou, A. Pandey, Life-cycle Assessment of Biorefineries, Elsevier, Amsterdam, Netherlands, 2017.
- [10] G. Thomassen, M. Van Dael, S. Van Passel, F. You, How to assess the potential of emerging green technologies? Towards a prospective environmental and technoeconomic assessment framework, Green Chem. 21 (18) (2019) 4868–4886.
- [11] O.I. Ögmundarson, S. Sukumara, M.J. Herrgard, P. Fantke, Combining environmental and economic performance for bioprocess optimization, Trends Biotechnol. 38 (11) (2020) 1203–1214.
- [12] Life Cycle Initiative, New! Hotspots Analysis methodological framework and guidance. https://www.lifecycleinitiative.org/new-hotspots-analysis-methodolo gical-framework-and-guidance/, 2017. (Accessed 4 April 2021).
- [13] G. Bravo, D. Lopez, M. Vasquez, A. Iriarte, Carbon footprint assessment of sweet cherry production: hotspots and improvement options, Pol. J. Environ. Stud. 26 (2) (2017) 559–566.
- [14] ISO 14044:2006 The International Standards Organization. Environmental Management — Life Cycle Assessment — Requirements and Guidelines, The International Organization for Standardization.
- [15] A. Corona, M.J. Biddy, D.R. Vardon, M. Birkved, M.Z. Hauschild, G.T. Beckham, G. C.O. National, Renewable Energy Lab, Life cycle assessment of adipic acid production from lignin, Green Chem. 20 (16) (2018).
- [16] S. Foteinis, A. Antoniadis-Gavriil, T. Tsoutsos, Life cycle assessment of algae-tobiodiesel shallow pond production systems in the Mediterranean: influence of

S.V. Obydenkova et al.

species, pond type, by(co)-product valorisation and electricity mix, Biofuels Bioprod. Bioref. 12 (4) (2018) 542–558.

- [17] D. Koch, M. Paul, S. Beisl, A. Friedl, B. Mihalyi, Life cycle assessment of a lignin nanoparticle biorefinery: decision support for its process development, J. Clean. Prod. 245 (2020).
- [18] O.g. Ólafur, S. Sumesh, L. Alexis, F. Peter, Environmental Hotspots of Lactic Acid Production Systems, GCB Bioenergy, 2020, pp. 19–38.
- [19] S. Bello, N. Pérez, J. Kiebist, K. Scheibner, M.a.I. Sánchez Ruiz, A. Serrano, A.n. T. Martínez, G. Feijoo, M.T. Moreira, Early-stage sustainability assessment of enzyme production in the framework of lignocellulosic biorefinery, J. Clean. Prod. 285 (2021).
- [20] A. Corona, R. Parajuli, M. Ambye-Jensen, M.Z. Hauschild, M. Birkved, Environmental screening of potential biomass for green biorefinery conversion, J. Clean. Prod. 189 (2018) 344–357.
- [21] P. Cheali, J.A. Posada, K.V. Gernaey, G.r. Sin, Economic risk analysis and critical comparison of optimal biorefinery concepts, Biofuels Bioprod. Bioref. 10 (4) (2016) 435–445.
- [22] K. Lan, L. Ou, S. Park, S.S. Kelley, B.C. English, T.E. Yu, J. Larson, Y. Yao, Techno-Economic Analysis of decentralized preprocessing systems for fast pyrolysis biorefineries with blended feedstocks in the southeastern United States, Renew. Sustain. Energy Rev. 143 (2021), 110881.
- [23] D. Humbird, R. Davis, L. Tao, C. Kinchin, D. Hsu, A. Aden, P. Schoen, J. Lukas, B. Olthof, M. Worley, D. Sexton, D. Dudgeon, in: Process Design and Economics for Biochemical Conversion of Lignocellulosic Biomass to Ethanol: Dilute-Acid Pretreatment and Enzymatic Hydrolysis of Corn Stover, National Renewable Energy Lab. (NREL), Golden, CO (United States), 2011, p. 147. Medium: ED; Size.
- [24] R. Davis, L. Tao, C. Scarlata, E.C.D. Tan, J. Ross, J. Lukas, D. Sexton, in: Process Design and Economics for the Conversion of Lignocellulosic Biomass to Hydrocarbons: Dilute-Acid and Enzymatic Deconstruction of Biomass to Sugars and Catalytic Conversion of Sugars to Hydrocarbons, National Renewable Energy Lab. (NREL), Golden, CO (United States), 2015, p. 133. Medium: ED; Size.
- [25] A. Giuliano, R. Cerulli, M. Poletto, G. Raiconi, D. Barletta, Optimization of a multiproduct lignocellulosic biorefinery using a MILP approximation, in: J. J. Klemeš, P.S. Varbanov, P.Y. Liew (Eds.), Computer Aided Chemical Engineering, Elsevier, 2014, pp. 1423–1428.
- [26] A. Giuliano, M. Poletto, D. Barletta, Process design of a multi-product lignocellulosic biorefinery, in: K.V. Gernaey, J.K. Huusom, R. Gani (Eds.), Computer Aided Chemical Engineering, Elsevier, 2015, pp. 1313–1318.
- [27] R. Shi, J.S. Guest, BioSTEAM-LCA: an integrated modeling framework for agile life cycle assessment of biorefineries under uncertainty, ACS Sustain. Chem. Eng. 8 (51) (2020) 18903–18914.
- [28] C.A. García-Velásquez, V. Aristizábal-Marulanda, C.A. Cardona, Analysis of bioenergy production at different levels of integration in energy-driven biorefineries, Clean Technol. Environ. Policy 20 (7) (2018) 1599–1613.
- [29] E. Martinez-Hernandez, G. Campbell, J. Sadhukhan, Economic value and environmental impact (EVEI) analysis of biorefinery systems, Chem. Eng. Res. Des. 91 (8) (2013) 1418–1426.
- [30] L. Tao, E.C.D. Tan, R. McCormick, M. Zhang, A. Aden, X. He, B.T. Zigler, Technoeconomic analysis and life-cycle assessment of cellulosic isobutanol and comparison with cellulosic ethanol and n-butanol, Biofuels Bioprod. Bioref. 8 (1) (2014) 30–48.
- [31] E.C.D. Tan, T.R. Hawkins, U. Lee, L. Tao, P.A. Meyer, M. Wang, T. Thompson, Biofuel options for marine applications: technoeconomic and life-cycle analyses, Environ. Sci. Technol. 55 (11) (2021) 7561–7570.
- [32] T.T. Manhongo, A. Chimphango, P. Thornley, M. Röder, Techno-economic and environmental evaluation of integrated mango waste biorefineries, J. Clean. Prod. 325 (2021), 129335.
- [33] Y. Cortes-Peña, D. Kumar, V. Singh, J.S. Guest, BioSTEAM, A fast and flexible platform for the design, simulation, and techno-economic analysis of biorefineries under uncertainty, ACS Sustain. Chem. Eng. 8 (8) (2020) 3302–3310.
- [34] S. Soltanian, M. Aghbashlo, S. Farzad, M. Tabatabaei, M. Mandegari, J.F. Görgens, Exergoeconomic analysis of lactic acid and power cogeneration from sugarcane residues through a biorefinery approach, Renew. Energy 143 (2019) 872–889.
- [35] S.V. Obydenkova, P.D. Kouris, D.M.J. Smeulders, M.D. Boot, Y. van der Meer, Lignocellulosic Multi-Product Biorefinery: Evaluation of Economic and Environmental Hotspots and Value Creation, EUBCE, 2021.

- [36] S.V. Obydenkova, P. Kouris, E.J.M. Hensen, H.J. Heeres, M.D. Boot, Environmental economics of lignin derived transport fuels, Bioresour. Technol. 243 (2017) 589–599.
- [37] IEA Bioenergy, Sustainable Lignin Valorization, 2021. https://www.ieabioenergy. com/wp-content/uploads/2021/11/Sustainable-Lignin-Valorization_rev22-11 -2021.pdf. (Accessed 20 January 2022).
- [38] Maersk, Sustainability report 2020. https://www.maersk.com/about/sustaina bility. (Accessed 11 July 2021).
- [39] P.D. Kouris, D.J.G.P. van Osch, G.J.W. Cremers, M.D. Boot, E.J.M. Hensen, Mild thermolytic solvolysis of technical lignins in polar organic solvents to a crude lignin oil, Sustain. Energy Fuels 4 (12) (2020) 6212–6226.
- [40] Chemelot InSciTe, Opening expanded multi purpose pilot plant. https://www.chemelot-inscite.com/en/news/opening-expanded-multi-purpose-pilot-plant, 2020.
- [41] S. Obydenkova, P.D. Kouris, E.J.M. Hensen, D.M.J. Smeulders, Y. van der Meer, M. D. Boot, Industrial lignin from 2G biorefineries assessment of availability and pricing strategies, Bioresour. Technol. 291 (2019).
- [42] S.V. Obydenkova, P.D. Kouris, D.M.J. Smeulders, M.D. Boot, Y. Meer, Modeling life-cycle inventory for multi-product biorefinery: tracking environmental burdens and evaluation of uncertainty caused by allocation procedure, Biofuels Bioprod. Bioref. 15 (5) (2021) 1281–1300.
- [43] GHG Emission Calculation Tool, 2021. https://ghgprotocol.org/calc ulation-tools#country_specific_tools_id. (Accessed 16 April 2021).
- [44] S. Suh, Handbook of Input-Output Economics in Industrial Ecology, Springer, Dordrecht ;, 2009.
- [45] R. Heijungs, S. Suh, The Computational Structure of Life Cycle Assessment, Kluwer Academic Publishers, Dordrecht ;, 2002.
- [46] H. Cai, J. Han, M. Wang, R. Davis, M. Biddy, E. Tan, Life-cycle analysis of integrated biorefineries with co-production of biofuels and bio-based chemicals: co-product handling methods and implications, Biofuels Bioprod. Bioref. 12 (5) (2018) 815–833.
- [47] C. Moretti, B. Corona, R. Hoefnagels, I. Vural-Gürsel, R. Gosselink, M. Junginger, Review of life cycle assessments of lignin and derived products : lessons learned, Sci. Total Environ. 770 (2021).
- [48] D.E. Garrett, Chemical Engineering Economics, Springer Netherlands, Dordrecht, 1989.
- [49] W.D. Seider, D.R. Lewin, J.D. Seader, S. Widagdo, R. Gani, K.M. Ng, in: Product and Process Design Principles : Synthesis, Analysis, and Evaluation, fourth ed., John Wiley & Sons Inc., New York, 2017.
- [50] K. Lokesh, A.S. Matharu, I.K. Kookos, D. Ladakis, A. Koutinas, P. Morone, J. Clark, Hybridised sustainability metrics for use in life cycle assessment of bio-based products: resource efficiency and circularity, Green Chem. 22 (3) (2020) 803–813.
- [51] C.L. Kinney, R. Gauche, What's in ISBL, OSBL, and the Factors? AACE Int. Trans. (2006) ES141–ES145.
- [52] R.C. Team, R: A Language and Environment for Statistical Computing, R Foundation for Statistical Computing, Vienna, Austria, 2020.
- [53] P. Bruijnincx, B. Weckhuysen, G.-J. Gruter, E. Engelen-Smeets, Lignin Valorisation : the Importance of a Full Value Chain Approach, Utrecht University, Utrecht, 2016.
- [54] M.-H. Cheng, H. Huang, B.S. Dien, V. Singh, The costs of sugar production from different feedstocks and processing technologies, Biofuels Bioprod. Bioref. 13 (3) (2019) 723–739.
- [55] T. Tobin, R. Gustafson, R. Bura, H.L. Gough, Integration of wastewater treatment into process design of lignocellulosic biorefineries for improved economic viability, Biotechnol. Biofuels 13 (1) (2020) 1–16.
- [56] DIRECTIVE (EU), 2018/2001 of the EUROPEAN PARLIAMENT and of the COUNCIL of 11 December 2018 on the Promotion of the Use of Energy from Renewable Sources (Recast) (Text with EEA Relevance), 2018.
- [57] P. Konstantin, M. Konstantin, Power and Energy Systems Engineering Economics : Best Practice Manual, Springer, Cham, Switzerland, 2018.
- [58] B. Nicole, T. Ian, A. Vivek, L. Yang, E. Shiva Zargar, S. Alyssa, P. Nathan, Comparing Sources and Analysis of Uncertainty in Consequential and Attributional Life Cycle Assessment: Review of Current Practice and Recommendations, The International Journal of Life Cycle Assessment, 2019, pp. 1–13.
- [59] C. Moretti, B. Corona, R. Edwards, M. Junginger, A. Moro, M. Rocco, L. Shen, Reviewing ISO compliant multifunctionality practices in environmental life cycle modeling, Energies 13 (14) (2020) 3579.