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Comparing biofuels through the lens of sustainability: A data envelopment analysis approach

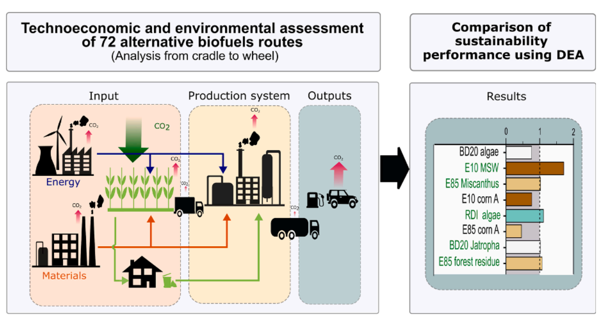
Richard Cabrera-Jiménez [a], Josep M. Mateo-Sanz [a], Jordi Gavaldà [a], Laureano Jiménez [a], Carlos Pozo [b,*]

[a] Departament d'Enginyeria Quimica, Universitat Rovira i Virgili, Av. Països Catalans 26, 43007 Tarragona, Spain
[b] Department of Chemical Engineering, Universitat Politècnica de Catalunya, EEBE, Av. Eduard Maristany 16, 08019 Barcelona, Spain

HIGHLIGHTS

- We compare the performance of 72 biofuel routes in terms of 12 sustainability indicators.
- DEA assigns an efficiency score to each biofuel, ranking them from best to worst.
- Renewable diesel was found the best fuel type, followed by biodiesel and ethanol.
- Waste biomass is preferred over lignocellulosic and 1st generation carbon sources.
- Benefits from demand-side measures are on pair with improving biofuel manufacturing.

GRAPHICAL ABSTRACT



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ABSTRACT

Liquid biofuels can facilitate the transition towards a more sustainable transportation sector by curbing carbon emissions while maintaining most of the current vehicle fleet. Today, a myriad of alternatives are available to produce biofuels, where different decisions for the fuel type, blend, conversion process and carbon source will affect the final cost and environmental impact of the product. In this contribution, we analyze the performance of 72 different biofuels routes based on 12 indicators that cover the three sustainability dimensions: economic, environmental and social. The proposed multi-criteria approach combines Data Envelopment Analysis with Life Cycle Assessment to evaluate biofuels from a cradle-to-wheel perspective, that is, considering the production chain spanning from biomass production to the combustion of the biofuel in the engine. Results reveal that there are 35 biofuels routes performing better than the rest, with renewable diesel being a better option than ethanol-based blends or biodiesel, and waste biomass preferred over cellulosic biomass or bio-oils. The selection of the

Abbreviations: B20, Diesel fuel with up to 20%v/v FAME content; CIDI, Compression Ignition Direct Injection; DEA, Data Envelopment Analysis; DMU, Decision Making Unit; E10, Gasoline fuel with up to 10%v/v bioethanol content; E85, Gasoline fuel with up to 85%v/v bioethanol content; FAME, Fatty Acid Methyl Ester; FWET, Freshwater ecotoxicity; FWEU, Freshwater eutrophication; GWP, Global Warming Potential; HT, Human ecotoxicity; HVO, Hydrotreated Vegetable Oil; LCA, Life Cycle Assessment; LCI, Life Cycle Inventory; LCIA, Life Cycle Impact Assessment; LO, Land Occupation; MSW, Municipal Solid Waste; NOx, Nitrogen Oxides; PMFP, Fine Particulate Matter Formation Potential; POFP, Photochemical Oxidant Formation Potential; RDI, Renewable Diesel Production Based on SuperCetane; RDII, Renewable Diesel Production Based on fluid catalytic cracker technology; SI, Spark Ignition; SBM, Slack Based Measure; TA, Terrestrial Acidification; TE, Terrestrial Ecotoxicity; VRS, Variable return to scale.

* Corresponding author.
E-mail address: carlos.pozo.fernandez@upc.edu (C. Pozo).

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carbon source proofed to be the most important decision, highlighting the need to consider regional aspects related to soil and climate before promoting a certain biofuel. Overall, our results can help to derive effective policies for the adoption of biofuels attaining the best performance at minimum cost and environmental risks.

1. Introduction

The continued growth of world population and the adoption of higher standards of living have risen energy demand to unprecedented levels. In the scenarios developed before the COVID-19 crisis, energy demand was projected to grow by 12% between 2019 and 2030 [1]. Among energy-consuming sectors, transport is the main player by the use of oil, covering 92% of fuel demand [2]. The widespread use of fossil fuels is the main anthropogenic source of greenhouse gases, responsible for climate change [3]. This evidences the fact that current practices for energy production are still far from sustainable [4], which raises concerns on the associated impacts in several environmental dimensions such as global warming, human health, land use or resource depletion [3–5].

In 2018, only 3.7% of fuel demand for transport was covered by renewable energy; with most of this being shouldered by biofuels (93%) and the rest provided by renewable electricity [6]. Biofuels such as biodiesel and bioethanol have been considered promising alternatives to fossil fuels for sustainable development due to their high potential to mitigate climate change [7–9]. Environmental pollution policies such as the Paris Agreement and the European Green Deal consider the widespread use of biofuels could importantly contribute to reaching reduction targets of 80–95% for greenhouse gas emissions by 2050 [10–12]. Many countries, e.g., the USA, Brazil, EU, China, have launched biofuel programs to reduce the use of fossil fuels in transport, and it is expected that the global share of biofuels in this sector will reach 17% by 2050 [6].

Biofuels refer to solid, liquid, and gaseous fuels that are produced from renewable biological sources. The most common biofuel is bioethanol, representing 82% of the total biofuel produced today [13]. Its main manufacturers are the United States and Brazil, with an annual production volume of 59.7 and 34.4 billion liters in 2020 and 2019, respectively [14–15]. The second most widely produced biofuel –and the most common in Europe [16]– is biodiesel, obtained by transesterification of oils or fats. Raw materials for biodiesel include vegetable oils, animal fats, and algae (third-generation biofuel), among others [17]. Bioethanol and biodiesel share the feature that can be used in internal combustion engines due to their high-octane number and high heat of vaporization [18], being both suitable either as an additive in gasoline blends or as pure fuels in modified engines.

Another relevant biofuel is renewable diesel (RD), sometimes called "second-generation biodiesel," "green diesel," or "HVO'' (hydrotreated vegetable oil) [19]. This biofuel is chemically similar to petroleum diesel (i.e., composed mainly of paraffins) but can be produced from a renewable feedstock containing triglycerides and fatty acids through various processes such as hydrotreating, gasification and pyrolysis [17]. Similar to biodiesel, its properties allow its use in conventional engines either as an additive or as a pure fuel [20–22].

In addition to curbing greenhouse gas emissions, the production of biofuels can offer other ancillary benefits to society [23]. On the one hand, it can diversify the supply of fuel to the transportation sector, providing a sustainable alternative to the existing transportation structure. On the other hand, it can also allow diversification of farmland while strengthening domestic agriculture by promoting biofuel feedstocks according to their geographical location and resource availability. In many cases, biofuels are suitable for current combustion engines and fuel stations, providing an interim solution before the required infrastructure for electric vehicles is in place. Note that, while electricity is the fastest-growing energy source in the transportation sector, it is projected to account for less than 2% of transport fuel consumption in 2050 in the United States [24]. Despite their advantages, biofuels are not exempt from negative side-effects, mainly related to the competition for land and water use [25].

As aforementioned, biofuels can be produced using different sources and processes, each generating different environmental impacts and achieving distinct performance in engines. In this context, the identification of the most convenient biofuels considering simultaneously the three sustainability pillars –economic, environmental and social– calls for multi-criteria decision-making tools (MCDM) [26]. The usefulness of such tools in solving environmental, socio-economic and technical barriers involved in energy planning has been widely acknowledged [27].

Different MCDM methods such as analytical hierarchy process [28], Multi-attribute value theory [29] and Data Envelopment Analysis (DEA) have been applied to assess different energy systems [30]. Amongst MCDM tools, we resort here to DEA, a non-parametric method for benchmarking alternatives [31]. The main advantage of DEA over other multi-criteria assessment methods is its capacity to combine multiple indicators into a single performance score, avoiding the need to define subjective weights between the indicators. This is very convenient in sustainability assessment as it allows to integrate indicators covering the three sustainability dimensions into a single metric, classifying alternatives as efficient or inefficient. In addition, DEA provides information on how much room for improvement is possible in inefficient alternatives compared to the best-performing processes.

During the last years, some authors have combined Life Cycle Assessment (LCA) with DEA to assess the overall level of sustainability of alternatives, enabling the identification of efficient processes with a focus on their sustainable performance. Examples of this combined application include liquid fuels production [32], electricity generation [33], bioenergy systems, [34], milk production [35], mussel cultivation [36], and grape production for vinification [37]. In the case of biofuels, previous works using DEA focused on particular features or echelons of the biofuel supply chain, e.g., cultivation locations [38], the biofuel production process [39], or the logistic network [40]. In other cases, the focus was put on a particular carbon source, would it be sugarcane [41] or algae [42], evaluating the complete supply chain of individual products such as bioethanol [43] and biodiesel [44]. While some of these works assessed the life cycle of biofuels, their scope covered, at most, stages up to the production of the fuel (cradle-to-tank), thus neglecting the combustion of the fuel during vehicle use (tank-to-wheel). Since this is the stage where most of the emissions take place and acknowledging that not all fuels show the same performance (in terms of emissions and energy efficiency) in vehicle engines, the inclusion of this stage in the analysis is crucial to obtain a holistic assessment of biofuels throughout their complete life cycle.

In this contribution, we evaluate the performance of 72 different routes for the production of biofuels considering the three sustainability dimensions, which are quantified here based on 12 different indicators. The 72 routes result from selected combinations of four biofuel blends, six possible fuels (i.e., ethanol, biodiesel, RD or HVO, diesel and gasoline) and 19 types of biological feedstocks. The analysis considers the whole life cycle of the biofuels, including cultivation, production, distribution, and final use of the fuel in combustion vehicles (i.e., cradle-to-wheel), everything quantified via LCA [45]. The resulting MCDM problem is solved with DEA [30] with the objective of evaluating and identifying the most suitable biofuel routes, which will be deemed efficient. For non-suitable biofuel routes, labelled as inefficient, we provide quantitative improvement targets that, if attained, would make them efficient. Finally, the presented contribution aims to provide a powerful framework for holistic assessments that could help policy-makers to develop better-informed regulations and achieve this way the emission reduction targets of current environmental policies for the transportation sector.

The remaining of this manuscript is structured in three sections as follows. Section 2 describes the methodology developed to evaluate biofuel production from a sustainability perspective and a cradle-to-wheel scope, paying special attention to DEA and its integration in the proposed framework. In Section 3, results are presented and analyzed in detail. Finally, in the conclusions, the implications for the technological, political and social spheres are discussed.

2. Methodology

The methodology used to assess the performance of biofuels consists of four main steps articulated around DEA, which is the cornerstone of our approach (Fig. 1). These steps are briefly summarized next, while further details are provided in the ensuing subsections.

Step 1 aims to obtain the data required to compute the indicators that will be used to assess the sustainability performance of the biofuels. This requires the collection of different types of data: from mass and energy balances for biofuel production processes, to traditional LCA data and complementary information such as costs.

With this information at hand, efficiency scores are computed for each biofuel using DEA in Step 2. To this end, each biofuel is modelled as a decision-making unit (DMU) in DEA and each sustainability indicator is classified as an input or an output to the DMU (further details in section 2.2). This analysis allows to classify biofuels as efficient (i.e., showing the best performance among alternatives) or inefficient (i.e., inferior to the best-observed practices). For the latter, DEA also provides improvement targets that, if attained, would make inefficient biofuels efficient.

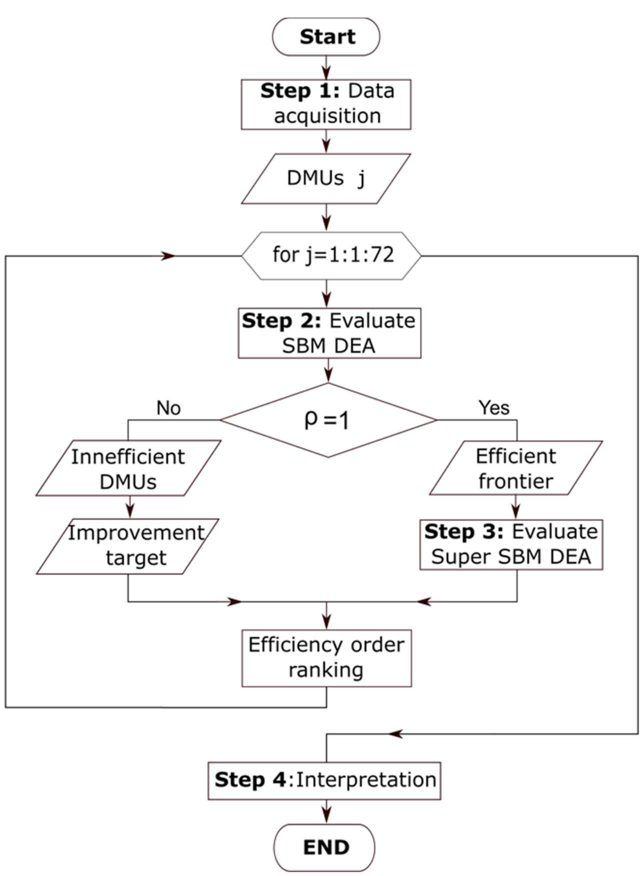

Fig. 1. Flowchart for the methodology proposed.

On the other hand, biofuels originally deemed efficient are further ranked in Step 3 by using a different DEA model based on a so-called super-efficiency score [46]. The combination of these results with the efficiency scores from Step 2 allows to build a sorted list from the best to the worst-performing biofuels that could aid policy-makers in developing effective regulations.

Finally, in Step 4, results are analyzed and interpreted considering the performance that selected biofuels could attain in different scenarios. Potential roadmaps for improvement are also discussed.

2.1. Data acquisition

The methodology described is used to compare the performance of 72 biofuel routes. This myriad of biofuel alternatives is obtained by combining selected options for the carbon source, the production process, the fuel type and the car engine where the biofuel will be used in (Fig. 2). Specifically, 19 types of biological feedstocks are considered as carbon sources; these cover lipids (i.e., vegetable oils, animal fats, and algae), cellulosic material (e.g., crop residues or woody biomass) and dedicated energy crops (e.g., sugarcane, maize) [47]. Regarding biofuel production processes, four types are studied: (i) fermentation of sugars (i.e., glucids) and (ii) biomass gasification to produce ethanol, (iii) transesterification of lipids (i.e., triglycerides) to produce FAME (fatty acid methyl esters), and (iv) hydrotreating of lipids (i.e., triglycerides) to produce hydrotreated vegetable oil (HVO) or renewable diesel (RD). In turn, each of the resulting biofuels can be blended differently to produce the final commercial fuel. In the case of bioethanol, two blends with gasoline are considered: E10, using 10% ethanol; and E85, using 85% ethanol. These blends are used in spark ignition (SI) engines. In the case of biodiesels, a blend consisting of 20% biodiesel-80% conventional diesel is assumed for use in compression ignition direct injection (CIDI) engines. Renewable diesel is obtained from two main processes, Super cetane (i.e., labelled here as RDI) and fluid catalytic cracker technology (i.e., named RDII) [47] and both are also used in CIDI engines but, in this case, as pure fuels as they are not blended. In all cases, engines are assumed to belong to a light vehicle carrying one single passenger.

Throughout this work, we use the term first-generation for biofuels derived from edible agricultural feedstock such as grain or sugars (e.g., corn, sorghum), the term cellulosic for biofuels produced from lignocellulosic biomass (e.g., willow, poplar) and the term bio-oil for biofuels obtained from oleaginous plants (e.g., soy, palm) [48–49].

Overall, 50 ethanol-based fuel routes are considered as follows. On the one hand, we study a total of 19 routes to produce ethanol though the fermentation of sugars from dedicated energy crops. Among these, 10 routes comprise the direct fermentation of biomass sugars. This is the case of five different production processes for corn, two processes combining usage of corn stover and corn, one process for sweet sorghum, one process for grain sorghum and one process for sugar cane. The remaining nine routes rely each on a different cellulosic carbon source and differ from the previous 10 in that the latter require a previous step consisting of an acid hydrolysis of the lignocellulose to produce simple sugars before these can be fermented into alcohol. Finally, six cellulosic carbon sources (i.e., six out of the nine cellulosic sources used) are employed to produce ethanol by biomass gasification. Overall, this yields a total of 25 routes to produce ethanol from biomass. Considering that the final ethanol product can be blended with gasoline in two different proportions (E10 and E85), this yields a total of 50 ethanol-based biofuel production routes.

Additional 22 routes based on bio-oil are also considered. Biodiesel can be produced from 8 additional carbon sources (in blue in Fig. 2) through transesterification of lipids. The resulting product is blended with diesel to form BD20. On the other hand, seven carbon sources can be used to produce RD using two main processes, Super cetane (i.e., RDI) and fluid catalytic cracker technology (i.e., RDII) [47]. These results in a total of 14 additional biofuels routes (note that RD is used as a stand-alone fuel, i.e., not blended with diesel).

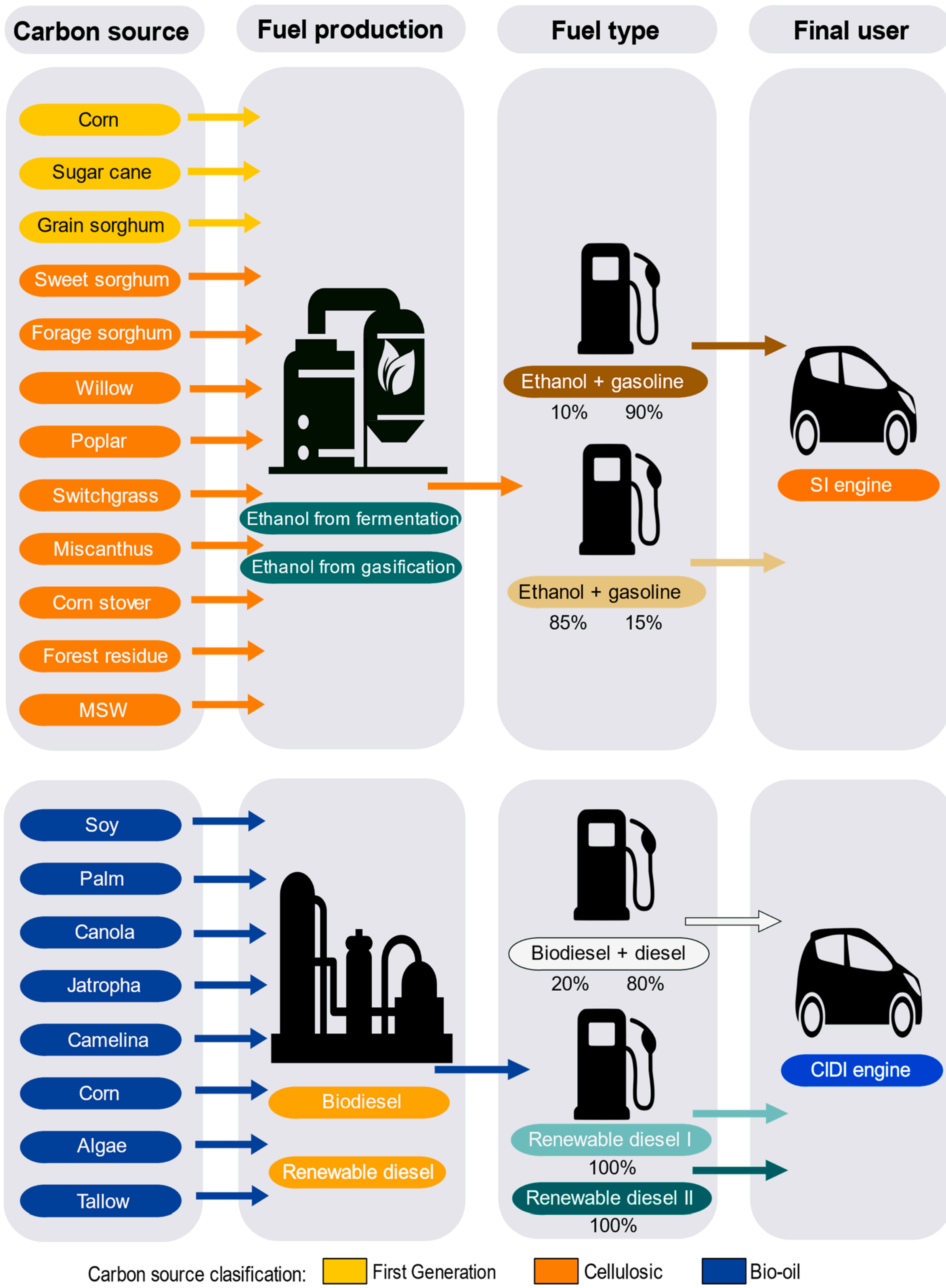

Fig. 2. Block-diagram providing the different alternatives considered as carbon source, fuel production process, blend and type of combustion engine. Carbon sources are depicted with a different color depending on whether they are first-generation (e.g., corn), cellulosic (e.g., poplar) or bio-oils (e.g., palm). SI: Spark Ignition; CIDI: Compression Ignition Direct Injection.

Overall, a total of 72 different biofuel routes are obtained: 50 for the case of ethanol that will be used in SI engines and 22 biofuels that will be used in CIDI engines.

For each of these 72 biofuel routes, 12 performance metrics covering the three sustainability dimensions are considered as follows. The economic dimension is assessed through the cost and the distance that can be travelled with the biofuel; the environmental dimension is evaluated through eight life-cycle impacts; and the performance in the social dimension is based on water use and land occupation since shortage of these resources can trigger social conflicts [50]. These performance metrics are assessed from a cradle-to-wheel perspective, thus accounting for all the resources and emissions occurring from cradle-to-tank (i.e., during the farming stage, biomass transportation and conversion to fuel) and from tank-to-wheel (i.e., combustion of the fuel in the vehicle engine).

The starting point for the calculation of the 12 indicators are the data collected from the GREET 2020 database [47], which provides information on the material and energy flows f (e.g., chemical reagents or electricity) required in each production stage p (i.e., cultivation, biomass transportation or biomass to fuel conversion) involved in the transformation of any carbon source into the corresponding fuel. These input flows, denoted here by $Input_{f,p}^{Raw}$ and reported in Tables S1-S9 in the Supplementary Material, are obtained for one liter of biofuel since this is the calculation basis selected in this contribution. Arguably, only a certain share of these inputs should be attributed to the requirements of biofuels themselves since other by-products are also obtained during the biofuel production process (e.g., corn-oil, electricity or glycerin). According to ISO 14040:2006 [51], allocation is the tool for partitioning input and output flows of a system between the product under study (e.g., biodiesel) and other by-products (e.g., glycerin). Among the different allocation methods available in the literature, here an economic allocation is used, as this is the baseline method for most LCA allocation situations [52]. The economic allocation generates an allocation factor (denoted here by AF , with $0 \leq AF \leq 1$) based on the quantity and the economic value of the biofuel itself and the corresponding by-products. This allows to compute the input f of stage p attributed to biofuel production ($Input_{f,p}$) as a certain share (AF) of the total input for the whole process ($Input_{f,p}^{Raw}$):

$$Input_{f,p} = Input_{f,p}^{Raw} AF \quad \forall f,p = \{farm, transport, conversion\} \tag{1}$$

Note that allocation only affects cradle-to-tank stages (i.e., farming, biomass transportation and conversion to fuel), since emissions incurred during combustion (i.e., tank-to-wheel) are solely attributable to the biofuel itself. The interested reader is referred to section 1.3 in the Supplementary Material for further details on the economic allocation.

With allocated input values available, these are next used to compute the different sustainability indicators. Calculations described next are repeated for every biofuel j, although subscript j has been dropped from equations and variables for simplicity. Since the GREET database builds upon United States (US) data, any complementary data used (e.g., power generation matrix) will also be based on the US to preserve data homogeneity, as suggested by Dyson. [53] .

First, the cost indicator ($Cost$) is computed as the summation of the product between the amount of input f required in every stage p (cradle-to-wheel) to produce one liter of the biofuel ($Input_{f,p}$) and the corresponding unitary costs (UC_f), as shown in Eq. (2). Unitary costs are obtained from different sources, as reported in Table S17.

$$Cost = \sum_{f,\ p \neq farming} Input_{f,\ p} UC_f \tag{2}$$

Note that the costs of farming inputs and extraction are neglected as they are assumed to be included in the cost of the vegetable oil feedstock.

The other economic indicator, i.e., the distance that can be travelled by burning the biofuel in the corresponding engine, is directly retrieved from GREET, as this information is readily available in the database.

As aforementioned, the environmental performance of the biofuel alternatives is quantified based on eight life-cycle impacts. Precisely, we use eight midpoint indicators of the ReCiPe approach following a hierarchical perspective and assuming allocation at the point of substitution. We choose midpoint over endpoint indicators as the former are considered less uncertain and, therefore, more reliable than the latter [54]. The indicators selected cover impacts related to human health (i.e., GWP, fine particulate matter formation, human ecotoxicity, photochemical oxidant formation potential) and ecosystems (i.e., terrestrial acidification, terrestrial ecotoxicity, freshwater eutrophication and freshwater ecotoxicity) [55]. The total impact in midpoint category u is computed by adding the corresponding impacts from the different life-cycle stages p ($Impact_{u,p}^{Stage}$), as shown in Eq. (3). For cradle-to-tank stages, impacts are computed as the product between the amount of input f required in the stage ($Input_{f,p}$) and the life-cycle impact in midpoint category u of producing a unit of input f ($Ecovector_{u,f}$) (see Eq. (4)). Ecovectors are obtained from Ecoinvent v3.7.1 database [56], using the activities reported in Table S18. For the combustion stage, direct emissions for different pollutants e ($Emission_{e,p}$), also provided by GREET, are converted into the corresponding impacts u by applying ReCiPe impact factors ($IF_{u,e}$, Eq. (5)) [47,57–58]. The results of this calculation (i.e., impacts for the combustion stage) are reported in Table S19 in the Supplementary Material.

$$Impact_u = \sum_p Impact_{u,p}^{Stage} \qquad \forall u \tag{3}$$

$$Impact_{u,p}^{Stage} = \sum_f Input_{f,p} Ecovector_{u,f} \qquad \forall u, p = \{farm, transport, conversion\} \tag{4}$$

$$Impact_{u,p}^{Stage} = \sum_e Emission_{e,p} IF_{u,e} \qquad \forall u, p = \{combustion\} \tag{5}$$

In the case of the GWP indicator, one final adjustment is required to account for the fact that, in the life cycle, emissions from biogenic carbon do not increase the total amount of carbon in the biosphere–atmosphere system. This is because biogenic carbon originates precisely by fixation of carbon from the CO_2 absorbed during photosynthesis, thus resulting in a net-zero cycle. Therefore, the CO_2 absorbed during biomass growth needs to be deducted from the total GWP obtained at the end of the fuel life-cycle to obtain the net balance of GHGs. To this end, the carbon content of the fuel is expressed in terms of carbon dioxide and discounted from the GWP obtained with Eq. (3). Carbon contents considered for the different fuels are 54.4 %w/w, 76.2 %w/w, 84.9 %w/w for ethanol, biodiesel, and renewable diesel, respectively [58], while fuel densities are provided in Table S21 in the Supplementary Material.

Finally, social indicators (i.e., land occupation and water use) are obtained as follows. The land occupation indicator ($Land$, in [$ha{\cdot}yr\ /l\ fuel$]) accounts for the annual land used for growing the necessary crops, neglecting land requirements for chemicals and energy production as these are expected to be significantly smaller than for harvesting biomass [56]. This indicator is computed from the amount of carbon feedstock needed to produce 1 L of biobased fuel ($Crop$, e.g., tons of poplar needed for 1 L of ethanol) and the annual yield of the corresponding crop ($Yield^{Crops}$, in tons of crop per hectare and year) (see Eq. (6)). Data for $Crop$ are obtained from GREET while the data for $Yield^{Crops}$ are reported in Table S25-S26, together with the corresponding data sources.

$$Land = \frac{Crop}{Yield^{Crops}} \tag{6}$$

In the case of the water use indicator ($Water$), two contributions are considered: the life-cycle water consumption for chemicals and energy production from cradle-to-wheel ($Water^{Inputs}$) plus the amount of water consumed for growing the corresponding crops ($Water^{Crops}$) (Eq. (7)). The former contribution is obtained by multiplying the amount of inputs ($Input_{f,p}$) by the life-cycle water consumption of producing one unit of such input (WC_f^{Inputs} , as retrieved from Ecoinvent for activities in Table S18)(Eq. (8)). On the other hand, the amount of water required to grow the corresponding crop can be calculated from Eq. (9), where land requirements are multiplied by the annual water consumption per hectare for the corresponding crop (LWC^{Crops} , in mm of water per square meter and year, see Tables S23-S24).

$$Water = Water^{Inputs} + Water^{Crops} \tag{7}$$

$$Water^{Inputs} = \sum_{f,p} Input_{f,p} WC_f^{Inputs} \quad (8)$$

$$Water^{Crops} = Land \cdot LWC^{Crops} \quad (9)$$

Note that values for LWC^{Crops} and $Yield^{Crops}$ correspond to agricultural land in conditions appropriate for the cultivation of each particular crop, with rainfall and artificial irrigation being both valid options to satisfy water requirements. If the performance of biofuel routes were to be evaluated for specific geographical areas, where annual rainfall is known, we suggest computing the water requirements based only on irrigation, as this can make a difference in the results obtained for the efficiency scores of routes based on certain crops (see Fig. S1 in Supplementary Material for further details). However, some of the considered crops (e.g., *Miscanthus,* switchgrass, poplar, willow) might be suitable for marginal land, thus avoiding competition with food at the expense of probably lower yields and larger water and chemical requirements.

In the case of materials that are considered residues or by-products of other crops (e.g., corn stover and forest residue), economic allocation factors of 15% [59] and 38% [60] respectively were applied for water use. The land required is obtained by multiplying the yield of these materials per hectare [t/ha] by the amount of feedstock needed to produce one liter of fuel.

The final values for the 12 indicators for the 72 biofuels will be referred to as the nominal values and are provided in Table S22 in the Supplementary Material and summarized here in Table 1, where biofuels are grouped into five categories according to their production process.

Note that we retrieved the data used to calculate indicator values from the same source for all biofuel routes to ensure a fair comparison between them. The only exception is farming data (i.e. water requirements and land yield for the different crops), which were retrieved from different sources, but always under the common assumption of adopting the most suitable conditions for growing each particular crop. Similarly, different production routes use different material inputs whose cost could not be retrieved from a single data source but were always determined by the corresponding commodity market. In addition, an uncertainty assessment was carried out to ensure reliable results and conclusions despite any potential data variation stemming from the occasional use of different sources or assumptions (see Section 2.5 for further details on this matter).

We next describe how indicator values are used in DEA to benchmark the sustainability performance of the different biofuels routes studied.

2.2. DEA fundamentals

DEA [31] is a data-oriented approach for evaluating the relative efficiency of a set of n similar entities called decision-making units (DMUs, indexed by j), which convert multiple inputs ($i = 1,\ldots,m$) into multiple outputs ($r = 1,\ldots,k$) [61]. Although DEA was originally devised to assess the productivity efficiency of production units, where inputs and outputs nomenclature was meaningful, later it has been widely used as a MCDM tool in any context. In the latter case, inputs and outputs can be any performance metric of interest, with the general agreement that inputs are metrics one is willing to minimize while outputs are metrics one seeks to maximize [61]. Some model variations also consider the potential existence of the so-called undesirable outputs, which are outputs to the production process one might want to reduce, e.g., polluting emissions [47].

In this contribution, each of the 72 biofuel route alternatives is modelled as a DMU whose relative performance is evaluated based on the 12 sustainability indicators described in the previous section and classified here as either inputs or outputs (desirable or undesirable, see Fig. 3). Dyson suggested that an appropriate discriminatory power could be achieved in DEA if the number of DMUs is at least 2·(m·k), where m·k is the product of the number of inputs times the number of outputs [53]; such a condition is satisfied in the present analysis (i.e., $72 > 2(3 \cdot 9)$).

For each DMU, DEA returns a performance score, also called efficiency score, lying between 0 and 1. DMUs (i.e., biofuels) with a score of 1 are referred to as efficient and are linearly combined to form the efficient frontier. Meanwhile, DMUs with a score strictly lower than 1 are considered inefficient and are projected onto the efficient frontier to generate the so-called virtual DMUs. Virtual DMUs can be understood as efficient versions of the projected DMU and allow the identification of the improvements that the inefficient DMUs should target to become efficient.

While these basic elements are common for all DEA approaches, a plethora of model variations has been put forward to date with the aim of better aligning model assumptions with the problem under study. Some of the modelling choices include the returns-to-scale (RTS), model orientation or the way in which the efficiency score is evaluated. These choices are explained in more detail in the following paragraphs.

The RTS aims to reflect whether DMUs operate or not at the same scale. The most common choices are the constant returns-to-scale (CRS), which assumes the ratio between inputs and outputs is constant regardless of the level of inputs, and the variable returns-to-scale (VRS), assuming a change in the inputs will produce a different change in the output depending on the input level [62].

Model orientation defines the way inefficient DMUs are projected

Table 1
Statistics of the sustainability indicators considered for the 72 biofuel routes. Values are for 1 L of fuel. Acronyms are provided in the table footnote.

Fuel type	BD20	E10	E85	RDI	RDII
Parameter	Median (min–max)	Median (min–max)	Median (min–max)	Median (min–max)	Median (min–max)
Cost [US$]	0.80 (0.74–1.12)	0.69 (0.68–0.74)	0.31 (0.21–0.75)	0.62 (0.41–1.17)	0.70 (0.46–1.30)
LO [m^2]	1.14 (0.04–3.65)	0.15 (0.01–0.28)	1.30 (0.001–2.36)	5.69 (0.20–11.04)	6.07 (0.22–11.67)
Water required [m^3]	0.54 (0.01–1.79)	0.16 (0.001–0.43)	1.32 (0.01–3.66)	2.09 (0.23–5.54)	3.46 (0.26–9.02)
GWP [kg CO_2-Eq]	2.52 (2.50–2.63)	2.51 (2.35–2.70)	0.85 (0.71–2.51)	0.59 (0.51–1.10)	0.31 (0.23–0.92)
FWET [10^{-2} kg 1,4-DCE]	0.96 (0.91–1.37)	0.56 (0.01–0.90)	1.23 (0.01–3.62)	0.80 (0.66–2.69)	0.86 (0.75–3.02)
FWEU [10^{-4} kg P-Eq]	0.45 (0.40–2.03)	0.44 (0.01–0.67)	0.85 (0.01–2.49)	0.64 (0.44–7.29)	0.70 (0.45–8.66)
HT [kg 1,4-DCE]	0.14 (0.14–0.29)	0.11 (0.09–0.15)	0.23 (0.10–0.49)	0.11 (0.10–0.75)	0.12 (0.10–0.88)
PMFP [10^{-3} kg PM10-Eq]	1.59 (1.53–2.33)	1.45 (0.002–1.78)	1.78 (0.001–3.21)	1.04 (0.74–4.10)	1.18 (0.86–4.84)
POFP [10^{-3} kg NMVOC]	5.31 (5.09–6.32)	4.36 (0.005–5.09)	5.64 (0.004–8.67)	4.00 (3.02–8.53)	4.40 (3.27–9.39)
TA [10^{-3} kg SO_2-Eq]	4.40 (4.28–5.45)	4.30 (0.005–5.23)	4.34 (0.004–7.91)	1.82 (1.28–6.15)	2.10 (1.50–7.29)
TE [10^{-3} kg 1,4-DCE]	2.58 (2.58–2.63)	0.67 (0.005–3.12)	4.58 (0.004–15.30)	2.45 (2.43–2.67)	2.47 (2.44–2.72)
Distance (km)	15.12	10.73	8.63	14.57	14.57

* BD20: Diesel fuel with up to 20 %v/v FAME content; E10: Gasoline fuel with up to 10 %v/v bioethanol content; E85: Gasoline fuel with up to 85 %v/v bioethanol content; RDI: Renewable Diesel Production Based on SuperCetane; RDII: Renewable Diesel Production Based on fluid catalytic cracker technology; LO: land occupation; Water: water used in farming plus water depletion produced during chemicals manufacturing; GWP: global warming potential; FWEU: freshwater eutrophication; FWET: freshwater ecotoxicity; HT: human ecotoxicity; PMFP: fine particulate matter formation; POFP: photochemical oxidant formation potential; TA: Terrestrial acidification; TE: terrestrial ecotoxicity.

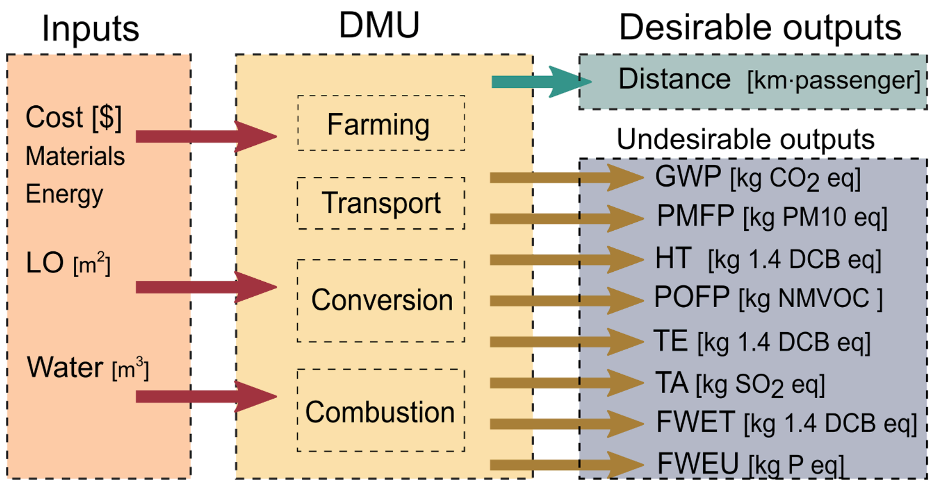

Fig. 3. Inputs and (desirable and undesirable) outputs considered for each biofuel (DMU). Units for each indicator are provided between brackets. LO: land occupation; Water: water used in farming plus water depletion; GWP: global warming potential; FWEU: freshwater eutrophication; FWET: freshwater ecotoxicity; HT: human ecotoxicity; PMFP: fine particulate matter formation; POFP: photochemical oxidant formation potential; TA: Terrestrial acidification; TE: terrestrial ecotoxicity.

onto the efficient frontier. In this regard, the most conventional alternatives are input-orientated, which attempts to minimize inputs while securing a certain level of output; and output-oriented models, where the opposite holds (i.e., outputs are expanded while maintaining the inputs at original levels). Non-oriented models, in which inputs and outputs are allowed to change simultaneously, are also widely used.

Finally, models are commonly grouped in two categories depending on whether the efficiency measure is radial or non-radial. Radial measures belong to the Debreu–Farrell measures and force changes in all the inputs (or all the outputs in an output-oriented model) to be proportional [63]. In contrast, non-radial measures belong to the Pareto–Koopmans measures [63] and allow inputs and outputs to vary in any possible way so that inefficient DMUs attain the efficient frontier. Examples of non-radial measures are Range Adjusted Measure, Russell Measure, Additive Model and Slack Based Measure (SBM) models. Note that not all possible model orientations can be used with any efficiency measure as these two choices are not always independent from each other. For instance, applying a non-radial model in cases where there is a linear dependence between inputs and outputs causes a loss of the original proportionality [64].

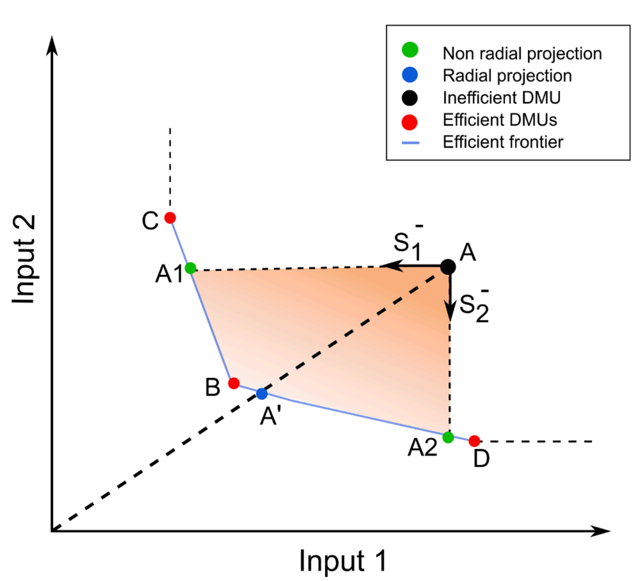

Fig. 4. Difference in projection onto efficient frontier by the radial and the non-radial SBM models.

Some of these concepts are illustrated in the following example (Fig. 4), where DEA is used to assess the efficiency of four DMUs (A, B, C, and D) against each other in a case considering two inputs and one output. If the output is dummy (e.g., all DMUs show the same performance in this output), DMUs can be represented in a two-dimensional cartesian plot as in Fig. 4. In this example, DEA would identify DMUs B, C, and D as efficient because there is no other DMU showing better performance, i.e., attaining lower inputs and/or higher output simultaneously. Efficient DMUs form the so-called efficient frontier, which corresponds to segment C-B-D when a VRS is considered, as in this example. Then, the model would project inefficient DMU A onto the efficient frontier to obtain the efficiency score and improvement targets for this unit. If the efficiency measure is radial and the model is input-oriented, then input 1 and input 2 would be decreased proportionally, yielding virtual DMU A'. In contrast, using a non-radial SBM model, the two inputs would be allowed to change non-proportionally. Indeed, Fig. 4 demonstrates this idea of non-proportionally wherein any projection in the quadrant A-A_1-A_2, as defined by slacks S_1^- and S_2^- (distance between the assessed and the virtual DMU) would be permitted in an SBM model. In this latter case, the virtual DMU of A could lie anywhere in the segment A_1-B-A_2, provided that inputs are not allowed to worsen.

DEA models based on non-radial measures are agreed to have a greater capacity to discriminate the DMUs under evaluation and yield a lower number of efficient units [65], therefore, being the preferred choice in environmental assessment. Among non-radial approaches, the most widely used one is the SBM model proposed by Tone [66], which, in its original formulation, treats undesirable outputs as inputs [67]. In our case, this translates into DMUs having 11 inputs (three original plus the eight undesirable outputs) and one output (the original desirable output). Previous studies have used this model to investigate issues related to water use relation with total factor productivity [68–69], the relation between energy use efficiency and either GDP [70] or economy development [71], the potential emission reductions and marginal abatement costs of energy-related CO_2 emissions [72], the measurement of environmental efficiency of transportation sector based on CO_2 emissions [73], and the relation between social fixed assets investment and GDP in the industry with SO_2 emissions [74]. The mathematical model is described in detail in the next section.

2.3. SBM non-oriented model

The SBM efficiency model proposed by Tone [75] is non-radial and computes the efficiency score based on the excess of inputs (s_i^- , which henceforth includes the original undesirable outputs) and the shortage of outputs (s_r^+). There are three variations of this model, i.e., input-oriented, output-oriented, and non-oriented, with the latter model referring to both input- and output-oriented. Working with the latter model prevents the need to decide between considering strong or weak disposability of environmental impacts, an assumption often made to deal with undesirable outputs [76]. Hence, without loss of generality, we use the non-oriented SBM model dealing with undesirable outputs as inputs for evaluating DMUs.

$$\rho^* = \min \frac{1 - \frac{1}{m}\sum_{i=1}^{m} \frac{s_i^-}{x_{i0}}}{1 + \frac{1}{k}\sum_{r=1}^{k} \frac{s_r^+}{y_{r0}}} \qquad \text{(m.1)}$$

$$\text{s.t.} \quad \sum_{j=1}^{n} \lambda_j x_{ij} + s_i^- = x_{i0} \qquad i = 1, 2, \cdots, m$$

$$\sum_{j=1}^{n} \lambda_j y_{rj} - s_r^+ = y_{r0} \qquad r = 1, 2, \cdots, k$$

$$s^- \geq 0, \quad s^+ \geq 0$$

$$\lambda_j \geq 0 \qquad j = 1, 2, \cdots, n$$

In this model, ρ is the SBM-efficiency score, x_{ij} is the value of input i of DMU j, y_{rj} is the value of output r of DMU j, and x_{io} and y_{ro} are the values of input i and output r of the DMU o under evaluation. In turn, s_i^- and s_r^+ are the input and output slacks, providing the distance from the DMU assessed to the efficient frontier. Slack variables in non-oriented SBM models provide information regarding the degree of inefficiency attained by each input and output individually [77].

This fractional programming problem can be transformed into a linear programming problem using the Charnes–Cooper transformation as follows:

$$\tau^* = \min t - \frac{1}{m} \sum_{i=1}^{m} \frac{S_i^-}{x_{i0}} \qquad \text{(m.2)}$$

$$\text{s.t.} \quad 1 = t + \frac{1}{k} \sum_{r=1}^{k} \frac{S_r^+}{y_{r0}}$$

$$\sum_{j=1}^{n} \Lambda_j X_{ij} + S_i^- = x_{i0} t \qquad i = 1, 2, \cdots, m$$

$$\sum_{j=1}^{n} \Lambda_j Y_{rj} - S_r^+ = y_{r0} t \qquad r = 1, 2, \cdots, k$$

$$S^- \geq 0, \ S^+ \geq 0, \ \Lambda \geq 0, \ t > 0$$

Note that the optimal solution of model (m.2) (e.g., $\tau^*, \Lambda^*, t^*, S^{-*}, S^{+*}$) can be used to derive the optimal solution of model (m.1) using the following relationships: $\rho^* = \tau^*, \lambda^* = \frac{\Lambda^*}{t^*}, S^{-*} = \frac{s^{-*}}{t^*}, S^{+*} = \frac{s^{+*}}{t^*}$.

2.4. Super-efficiency

DEA evaluates the relative efficiency of DMUs but does not further rank among the efficient units. This sometimes results in DEA providing a long list of promising (efficient) alternatives, upon which decision-makers need to choose based on additional criteria. To provide more accurate rankings without having to resort to additional considerations, the super-efficiency score has become an option to discriminate between efficient DMUs.

Super-efficiency models identify the best-performing DMUs by assigning an efficiency score greater than one, thus facilitating comparison with rankings based on parametric methods [46]. These models execute standard DEA models under the assumption that the DMU assessed is excluded from the efficient frontier. In other words, in super-efficiency DEA models, the virtual DMU must be constructed using the remaining DMUs only [78]. For the case of the SBM model m.1, one can resort to the super-SBM model proposed by Tone [66] for evaluating efficient DMUs ($\rho^* = 1, S^- = 0, S^+ = 0$) . The model formulation is as follows:

$$\delta^* = min \frac{\frac{1}{m}\sum_{i=1}^{m} \frac{\bar{x}_i}{x_{i0}}}{\frac{1}{k}\sum_{r=1}^{k} \frac{\bar{y}_r}{y_{r0}}} \qquad \text{(m.3)}$$

$$\text{s.t.} \quad \bar{x} \geq \sum_{j=1, \neq 0}^{n} \lambda_j x_j$$

$$\bar{y} \leq \sum_{j=1, \neq 0}^{n} \lambda_j y_j$$

$$\bar{x} \geq x_0, \bar{y} \leq y_0, \lambda \geq 0$$

The previous SBM model (m.1) and the super SBM model (m.3) selected for this work assume constant returns to scale (CRS), although these models could be extended to variable returns to scale (VRS) by adding equations $\sum_{j=1}^{n} \lambda_j = 1$ and $\sum_{j=1, \neq 0}^{n} \lambda_j = 1$ in models (m.1) and (m.3), respectively.

2.5. Dealing with data uncertainty in DEA

Regardless of the efforts invested in collecting data with the highest quality, DEA results might always be affected by data inaccuracies or simplifications, which could lead to spurious efficiency scores and rankings. To overcome this, we consider uncertainty in our data in an attempt to obtain more robust results and conclusions under different potential realizations of the uncertainty. Without loss of generality, we assume each indicator follows a uniform distribution spanning $\pm 10\%$ of its nominal value. These distributions are then discretized into 100 different scenarios for each DMU using Monte Carlos sampling. Finally, following the approach of Ewertowska et al. [79] 100 independent DEAs (i.e., one for each scenario) are solved, in addition to the nominal scenario, yielding a distribution of efficiency scores for each DMU (rather than a single value).

3. Results

The non-oriented SBM efficiency model (m.2) and the non-oriented SBM super-efficiency approach (m.3) were coded in GAMS v32.1.0 [80] and solved in an AMD Ryzen 5 4500U processor for each of the 72 DMUs in each scenario. Each instance took less than 1 s of CPU time to be solved. The results obtained are described next, starting with the efficiency and super-efficiency scores, then moving to the analysis of inefficient alternatives and finally exploring different improvement scenarios for the transportation sector. For the sake of simplicity, the discussion will focus on the results obtained for the nominal scenario, except for cases where result distributions are explicitly mentioned.

3.1. Efficiency assessment

Fig. 5 provides the combined results for the efficiency and super-efficiency DEAs, with inefficient biofuels being represented based on their efficiency score and efficient biofuels depicted based on their super-efficiency score. Specifically, horizontal bars provide the efficiency score in the nominal scenario, while overlapped boxplots provide information on the distribution of the efficiency scores obtained in the remaining 100 scenarios. Results reveal that 48% of the 72 biofuels routes analyzed are efficient in the nominal case (Fig. 5a), meaning that there is no other biofuel showing superior performance in all the

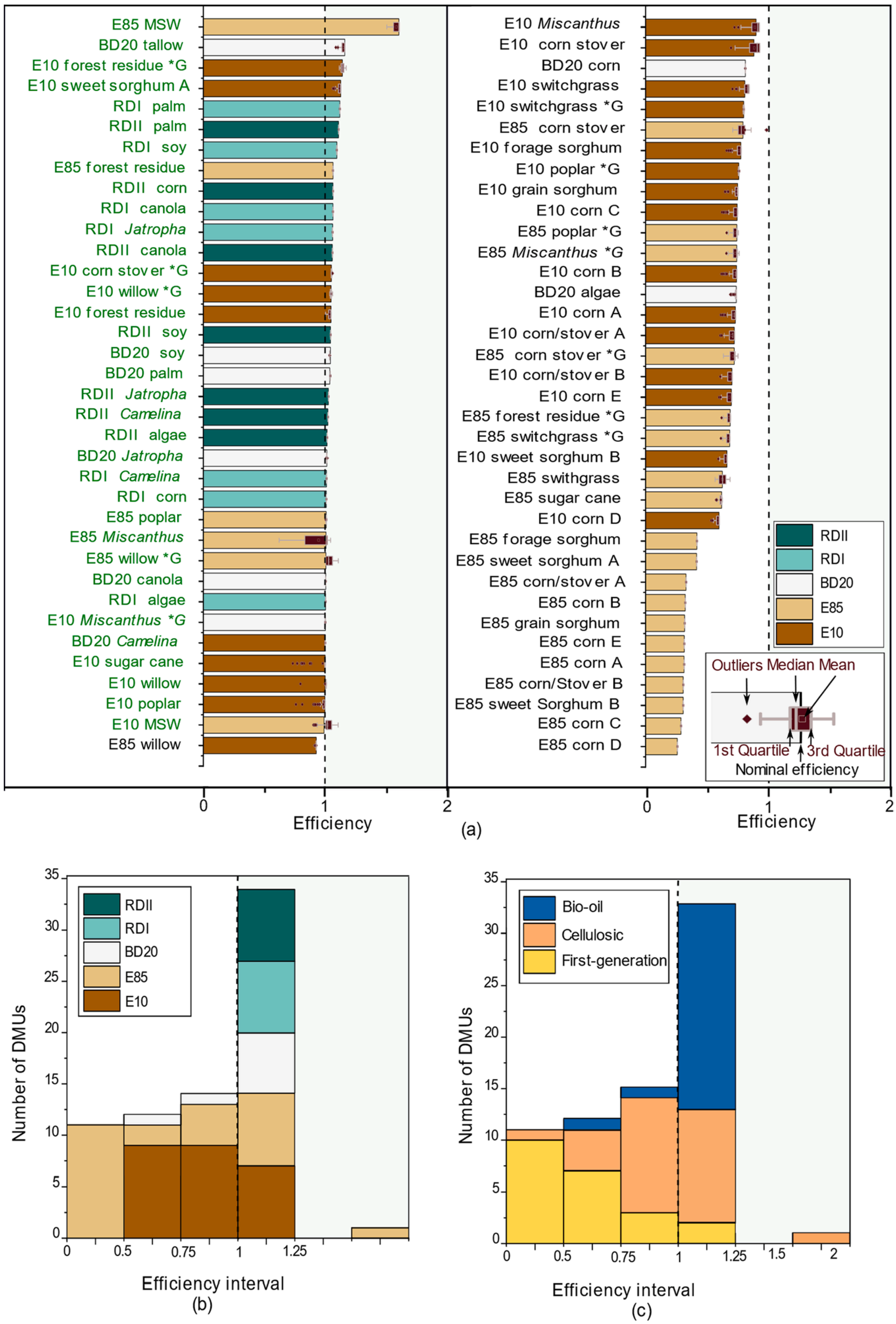

Fig. 5. Efficiency scores for biofuels. (Super)efficiency scores for the 72 biofuels routes are provided as horizontal bars in subplot (a), with biofuels sorted in decreasing order of efficiency and efficient biofuels depicted with a green label. Histograms at the bottom of the figure group results per type of biofuel (subplot (b)) or type of feedstock (subplot (c)). ETOH corn A: Dry mill corn without oil extraction; ETOH corn B: Dry mill corn with oil extraction; ETOH corn C: Wet milling corn; ETOH corn D: combined dry and wet milling corn; ETOH corn/stover A: integrated corn/stover ethanol (associated with corn); ETOH corn/stover B: integrated corn/stover ethanol (associated with stover); ETOH corn E: Gen dry milling corn with oil extraction; ETOH sweet sorghum A: Conventional; ETOH sweet sorghum B: Integrated.*G: Ethanol produced by gasification. BD20: Diesel fuel with up to 20 %v/v FAME content; E10: Gasoline fuel with up to 10 %v/v bioethanol content; E85: Gasoline fuel with up to 85 %v/v bioethanol content; RDI: Renewable Diesel Production Based on SuperCetane; RDII: Renewable Diesel Production Based on fluid catalytic cracker technology. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

sustainability indicators simultaneously. This implies there is a pool of 35 biofuels from which policy-makers can select the most suitable alternatives to promote according to the regional context (e.g., land availability, farmer preferences or the most abundant type of vehicle -SI vs. CIDI-).

The highest efficiency score, standing at 1.61, is achieved by the blend using 85% of ethanol from municipal waste, owning to different factors. On the one hand, low cost, water and land occupation requirements are allocated to MSW compared to other feedstocks (e.g., 0.01 m^3 of water/liter of E85 from MSW, compared to 0.76 m^3 of water/liter of E85 from dry mill corn without oil extraction). On the other hand, this is also attributable to the production process itself, which takes advantage of low-cost fermentable sugar sources. In the case of MSW, the energy demand of the process is self-satisfied by using either a fraction of the biomass feedstock or the residues from the fermented biomass, also exporting any surplus of energy that might be produced. This makes the fossil carbon emissions, as well as the impacts associated with energy generation and transportation, lower for MSW than for any first-generation biomass. Indeed, the production of 1 L of ethanol from first-generation biomass (i.e., fermentation of simple sugars) emits on average 0.47 kg CO_2 eq, while the production of 1 L of ethanol from cellulosic materials and an acid hydrolysis process emits only 0.21 kg CO_2 eq.

Interestingly, the blend using 10% ethanol from MSW shows a modest efficiency in the nominal scenario (1.00) and even has an 11% chance of being inefficient. This inferior result compared to the E85 blend stems from the increased amount of poor-performing gasoline present in the blend. Still E10 from MSW can achieve efficiencies as high as 1.11 in some scenarios; this would place it as the eighth fuel if sorted according to the maximum efficiency score displayed in any scenario.

Despite the promising results of biofuels based on MSW, the availability of waste suitable for biofuel production could limit the displacement of fossil fuels with these alternatives. As an example, 0.23 kg of dry MSW is generated per day and person in Europe. If all this waste were used to produce E85, 0.08 L would be obtained, yet this would only cover 1.8% of the daily per capita demand for fuel in the region (i.e., 4.4 L/day person) [81–82].

We next turn our attention to the lowest efficiency score in the nominal scenario (0.26), which corresponds to ethanol from corn (i.e., E85 from combined dry and wet milling corn). This can be explained by its high resource requirements and low mileage achieved per liter of biofuel (i.e., 8.63 km compared to 14.57 km in the case of any renewable diesel).

Comparing the five different types of fuels studied (i.e., E10, E85, BD20, RDI, RDII), it is observed that there is at least one efficient biofuel for each of them in the nominal scenario (Fig. 5b). This does not mean that all fuel types performed equally well: whilst almost all the BD20, RDI and RDII fuels are found efficient, only 30% of ethanol-based fuels (15 out of 50) achieve the efficient status (Fig. 5a). This indicates that the fuel type alone is not enough to draw strong conclusions, and that the carbon source should also be explored.

To this end, we classify biofuels into three groups depending on their carbon source: (i) bio-oils, consisting of animal fat and vegetable oil; (ii) cellulosic material, i.e., those made of lignocellulosic biomass; and (iii) first-generation sources, including sugars and starch (Fig. 5c). Again, we find examples of efficient biofuels for any type of carbon source, yet some patterns can still be observed. Bio-oil-based fuels (e.g., BD20, RDI, RDII) show, on average, the highest efficiency scores, standing at 1.03, compared to 0.91 for biofuels based on lignocellulosic biomass and 0.56 for those based on first-generation biomass. This also translates into a larger share of bio-oil-based fuels deemed efficient: 90%, compared to 46% in the case of biofuels from lignocellulosic feedstock and 59% for those based on first-generation biomass. These results are explained by the lower fuel consumption in CIDI engines compared to SI engines and, in the case of renewable diesel, by the possibility of using 100% bio-based fuels (i.e., no blends) without affecting the engine performance [83–85]. Inspection of data (see Table 1) reveals that fuels used in SI engines have 15% lower median prices than the fuels used in CIDI engines, 0.67$/liter vs. 0.78$/liter. This difference is reversed when the comparison considers fuel consumption per km, where CIDI-type fuels achieve a lower price (0.054$/km vs. 0.07$/km). Therefore, while renewable diesel generates greater environmental impacts per liter of fuel burned (see Table S19 and S20), this is offset by the achievement of longer distances travelled, which ultimately translate into lower impacts per km (i.e., lower inputs for the same level of output).

Overall, these results call for encouraging the use of renewable diesel over traditional biodiesel or bioethanol owing to their lower GWP in the life cycle, their lower fuel consumption rate, and their lower exhaust particle emissions per km [86]. In cases where biodiesel is still to be used, cellulosic carbon sources are preferred over first-generation biomass; this might also avoid concerns about competition with food by growing crops in marginal land. In addition, the processes for converting cellulosic biomass into bioethanol typically devote part of the biomass feedstock to the cogeneration of heat and electricity for self-consumption. This not only reduces the input requirements allocated to the biofuel, but also lowers the dependence on the domestic electricity mix by satisfying part of the energy demand of the process through renewable sources (~74% on average) [47]. Cellulosic materials are currently becoming more competitive, achieving better performance and lower cost thanks to advances in the production of enzymes for the degradation of lignocellulosic materials into simple fermentable sugars (e.g., pentoses, hexoses) [87]. However, replacing the total diesel consumption in Europe (i.e., 287Mtoe [88]) with renewable diesel from canola would require exploiting 90% of the total agricultural land available in the region (1.15 million km^2 [89]), clearly an unrealistic scenario.

Inefficient units are mostly based on corn and sorghum grains, also part of first-generation ethanol blends. Their low performance is due to different factors. On the one hand, the feedstock costs are higher for these fuels than for lignocellulosic materials (e.g., 130$/t or 350$/t of corn and sorghum, respectively, compared to 58$/t for *Miscanthus,* as an example of lignocellulosic material) [90]. Besides, environmental impacts generated during the farming stage of corn and sorghum are more significant owing to the higher use of machinery, transportation, pesticides and fertilizers (e.g., 194 g of fertilizer per liter of corn-based ethanol, compared to 10 g of fertilizer per liter of willow-based ethanol, see Tables S1 and S5).

One aspect that stands out is the low efficiency of fuels based on *Jatropha* compared to soybeans, even though the former has a higher oil content, lower water requirements, and lower land occupation (see Table S28). This might be due to the three times higher energy requirement for the farming stage per kg of feedstock compared to soybeans.

Inefficient biofuels based on bio-oil correspond to those coming from corn and algae sources. This is not only due to the carbon source but rather to the need to mix these fuels with fossil diesel. Indeed, corn and algae are efficient when they are used to produce a fuel based on 100% renewable carbon (RDI and RDII), allowing for the reduction of carbon emissions and other environmental impacts associated.

It is also observed that data uncertainty has a marginal role in shaping efficiency scores, at least to the extent of affecting the trends observed. Most of the biofuels are efficient or inefficient in all the scenarios and the nominal case, with only five biofuel routes changing depending on the realization of the uncertainty. These are E85 *Miscanthus* (with a 74% chance of being efficient), E10 sugar cane (90% chance), E10 willow (90% chance), E10 poplar (60% chance) and E10 MSW (89% chance). Among them, only the aforementioned E10 from poplar and E85 from *Miscanthus* show their performance clearly affected (efficiency score between 0.76–1.00 for the former and 0.62–1.05 for the latter).

Given that biofuels are mainly considered a potential solution for the climatic problem, and acknowledging that other environmental impacts are also important, we next explore in detail the performance achieved by some biofuels in terms of their GWP (Fig. 6a). It is observed that the combustion stage is the one that contributes the most to this impact category, being responsible for 80% of carbon emissions on average. However, part of these emissions would come from biogenic carbon, which does not contribute towards the GWP because it does not increase the total amount of carbon in the biosphere–atmosphere system in the life cycle. The share of emissions stemming from biogenic carbon depends on the fuel and the carbon source used, and can be as high as 93% of combustion emissions for renewable diesel made from palm (i.e., 93% of total GWP without deducting biogenic CO_2). In this particular example, subtracting biogenic emissions would place the combustion stage at 53% of the total GWP of the fuel. In contrast, biogenic emissions are low for biofuels based on blends with gasoline (e.g., E10 from willow) or with conventional diesel (e.g., BD20 from palm), where even after discounting biogenic emissions, the combustion stage still represents 82% and 81%, respectively, of the cradle-to-wheel GWP. Overall, this suggests that blends with low biofuel content (e.g., 20%) will have a limited benefit on climate change, which calls for policies promoting pure biofuels or blends with higher biofuel shares.

A totally different picture emerges when other environmental impacts are assessed. In the case of terrestrial ecotoxicity (Fig. 6b), the production stage contributing the most towards the total impact depends strongly on the fuel type and carbon source. For biofuels based on bio-oil, the combustion stage contributes 33% of total terrestrial ecotoxicity, while for biofuels based on ethanol, combustion emissions represent only 1% of the total terrestrial ecotoxicity. Indeed, the emissions of polycyclic aromatic hydrocarbons (PAHs) released during biofuel combustion in a vehicle engine significantly affect the difference in terrestrial ecotoxicity between the two fuel types. Note that, in the absence of more specific data, we only differentiate PAH emissions between the two types of engines considered (CIDI vs SI), but not between different blends used in the same engine. This assumption is based on the observation that PAH emissions are mostly dictated by the engine operating conditions [91].

In the case of third-generation biofuels, i.e., those using algae as feedstock, the stage where algae is converted to renewable diesel is highly energy-intensive, mainly due to the oil extraction process. This makes this stage the most important in terms of terrestrial ecotoxicity (70%) and the second most important in GWP (25%) for algae-based renewable diesel, and therefore could be object of further research aiming at improving the sustainability level of these biofuels.

The results obtained through the methodology applied in this contribution for the evaluation of biofuels are in agreement with those found using other metrics such as RepSIM [92]. Despite differences between the two approaches exist, both methodologies combine economic, environmental and social indicators to perform a holistic sustainability assessment, finding that the most sustainable alternatives result from using low-value waste products as carbon sources (i. e., MSW, tallow) and processes that involve cracking energy dense molecules and reforming them in the presence of hydrogen (e.g., HVO or Fischer-Tropsh).

3.2. Inefficiency assessment

Once identified, inefficient units are projected onto the efficient frontier, and improvement targets are computed for their different sustainability indicators. These improvement targets are provided per DMU in Fig. 7 as the median percentual changes required with respect to the nominal values across the 100 different scenarios (i.e., decrease for inputs and undesirable outputs, and increase for outputs). In the interest of clarity, the information for the 37 inefficient DMUs is lumped, here, into ten groups with similar carbon sources; the complete results are provided in Table S32 in the Supplementary Material, while detailed results for the nominal scenario are given in Table S31.

Most inefficient units need to achieve significant reductions in land occupation, water use, and terrestrial ecotoxicity to become efficient. This is especially evident in the case of E85 fuels due to the higher fuel consumption of SI engines, causing, in turn, the increase of impacts from fuel production for the same mileage.

Corn-based E10 requires improvements in all the inputs, with the largest reductions observed in water use (96%), terrestrial ecotoxicity (73%), land occupation (63%) and freshwater eutrophication (27%). This poor performance is mainly due to two factors. On the one hand, corn farming is a very demanding process, requiring significant amounts of land, water and energy compared to other crops (e.g., on average, 200% more energy than for cellulosic materials such as *Miscanthus* or poplar). On the other hand, the conventional conversion process from

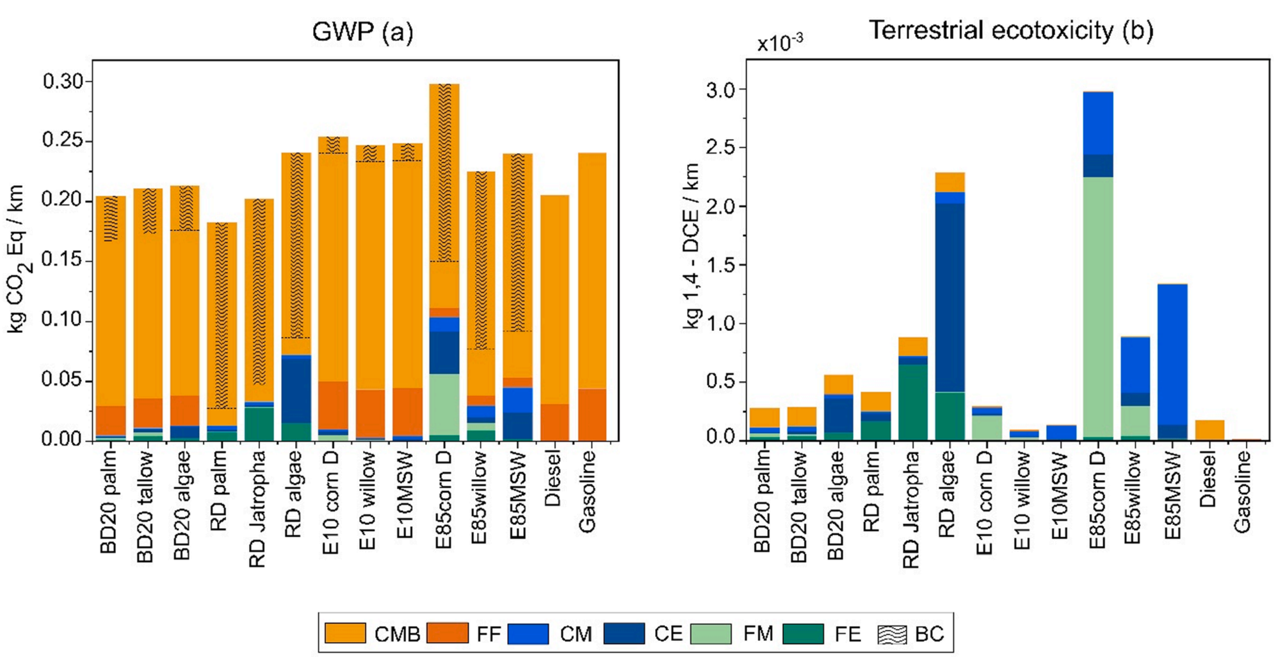

Fig. 6. Breakdown of the GWP (subplot (a)) and terrestrial ecotoxicity (subplot (b)) generated in the life cycle of selected biofuels. CMB: Combustion; FF: Fossil fuel production; CM: Materials for the biofuel production; CE: Energy for biofuel production; FM: Farming materials; FE: Farming energy; BC: Biogenic Carbon.

		Inputs			Undesirable Outputs								Outputs
DMU	Eff.	Cost	LO	Water	GWP	FWEC	FWEU	HT	PMFP	POFP	TA	TE	Distance
BD20 algae	0.75	15%	35%	0%	8%	0%	61%	44%	18%	18%	0%	74%	0%
BD20 corn	0.82	0%	34%	72%	15%	0%	5%	7%	18%	18%	14%	9%	0%
E10 cellulosic	0.72	0%	74%	55%	1%	10%	5%	4%	1%	4%	1%	50%	0%
E10 cellulosic sorghum	0.78	6%	75%	83%	0%	8%	14%	11%	3%	7%	1%	79%	0%
E10 corn	0.76	1%	63%	96%	10%	13%	27%	20%	6%	8%	2%	73%	0%
E10 grain sorghum	0.73	0%	87%	27%	6%	18%	23%	20%	4%	5%	0%	70%	0%
E85 cellulosic	0.31	0%	0%	54%	29%	36%	49%	44%	31%	32%	49%	77%	0%
E85 cellulosic sorghum	0.63	41%	99%	99%	39%	40%	67%	68%	52%	63%	45%	98%	0%
E85 corn	0.35	27%	99%	98%	62%	57%	74%	74%	57%	62%	46%	99%	2%
E85 sugar cane	0.64	0%	0%	77%	20%	0%	53%	59%	40%	49%	41%	72%	0%
Average improvement		11%	59%	57%	12%	14%	28%	25%	17%	20%	13%	58%	1%

% decrease	0	20	40	60	80	90	100
% increase	0	20	40	60	80	90	100

Fig. 7. Median improvement targets for inefficient biofuels to become efficient considering 100 possible scenarios. E10 cellulosic: median improvement targets across E10 switchgrass, E10 Miscanthus & E10 corn stover; E10 cellulosic sorghum: median improvement targets across E10 sweet sorghum B & E10 forage sorghum; E10 corn: median improvement targets across E10 corn A-B-C-D-E & E10 corn/stover A-B; E85 cellulosic: median improvement targets across E85 switchgrass & E85 corn stover; E85 cellulosic sorghum: median improvement targets across E85 sweet sorghum A-B & E85 forage sorghum; E85 corn: median improvement targets across E85 corn A-B-C-D-E & E85 corn/stover A-B; LO: land occupation; Water: water used in farming and water depletion; GWP: global warming potential; FWEU: freshwater eutrophication; FWET: freshwater ecotoxicity; HT: human ecotoxicity; PMFP: fine particulate matter formation; POFP: photochemical oxidant formation potential; TA: Terrestrial acidification; TE: terrestrial ecotoxicity.

first-generation biomass to E10 covers all its energy demand with the domestic energy matrix, being exposed to the cost and impacts of the country mix. This is a clear disadvantage compared to the conversion process for cellulosic feedstocks, which satisfy part of their energy demand by using a certain share of the biomass feedstock to generate heat and electricity for self-consumption.

The higher ethanol content of corn-based E85 increases the resources needed for crop farming, which in turn raises the improvements required for water use (94%), land occupation (95%) and terrestrial ecotoxicity (98%) to levels hardly achievable. Indeed, the first two indicators, clearly associated with farming of the grain, seem already unattainable. Even if irrigation could be fully covered by rainfall in certain regions, meeting the improvement target for land occupation would entail almost doubling the yield: from the current 17.31 t/ha/yr (Table S25) up to a target yield of 34.27 t/ha/yr. On the other hand, fertilizers and pesticides used during farming have a significant contribution to terrestrial ecotoxicity (Fig. 6b), and again it is challenging to imagine that magnitude of reduction without affecting a crop yield that should be further improved. In addition, E85 shows the lowest mileage per liter of fuel used (i.e., higher fuel consumption per kilometer), which result in higher emissions from combustion and, therefore, higher reductions in human ecotoxicity (74%), freshwater eutrophication (74%), photochemical oxidant formation potential (57%), GWP (62%), freshwater ecotoxicity (57%), fine particulate matter formation (57%) and terrestrial acidification (46%). Furthermore, the mileage achieved should be improved by 2%; this could be pursued by using engines built to work with ethanol-blends (flex-fuels vehicles) or by incorporating turbochargers [93].

In the case of E10 from cellulosic feedstocks, reductions are required in most inputs, yet these are more modest compared to E10 based on first-generation biomass. The reason is that farming of cellulosic feedstock requires less energy and materials than farming of first-generation biomass does, thus penalizing the contribution of biomass production for the latter. The most important improvements requested for E10 based on cellulosic biomass are reductions of 55% and 74% in water requirements and land occupation, respectively. The former impacts are mainly caused by the use and production of fertilizers, leaving rotational crops and organic fertilizers as the most promising option for their abatement [94,95]. On the other hand, impacts on land occupation might imply a yield increase that could be pursued by growing crops in best-endowed regions, i.e., on soils with adequate natural moisture available and non-winter climates [96]. Works by Castillo et al. [97] and Zhang et al. [98]offer a suitability analysis of soils for different crops (e.g., *Miscanthus*, switchgrass, poplar, *Jatropha*).

The production process for E10 and E85 based on cellulosic sorghum (i.e., sweet and forage sorghum) is the same as for the other cellulosic feedstocks, devoting part of the biomass to satisfy its own energy requirements. Despite this, more demanding improvements are found when using sweet, and forage sorghum since growing these crops entails higher costs and water requirements than other cellulosic feedstocks, making them a poorer choice.

The inputs requiring the highest reductions for corn-based BD20 are water use (72%) and land occupation (34%). Inefficiencies in these categories are due to the low oil content of the corn grain (about 3–4%), which results in larger feedstock requirements even after the economic allocation (i.e., only 17% of the inputs for corn production are allocated to the biodiesel). An energy sector with a strong dependence on biomass might alleviate global warming at the expense of imposing additional burdens on land or freshwater use; however, the urgency to solve the climatic problem and the fact that land-system and freshwater use planetary boundaries are not yet transgressed might fully justify the transition [99,100].

Algae-based BD20 shows high oil content (up to 35% in dry weight) and low water requirements, which do not prevent it from needing important improvements in human toxicity (44%) and land occupation (35%). These two impacts are directly related to the energy needs for algae drying and oil extraction, so advances in energy efficiency and the oil extraction process, such as supercritical fluid extraction [101], could reduce the existing gap between the current and the target performance. In addition, a reduction of 74% is required for terrestrial ecotoxicity.

Impacts in this category are generated mainly during diesel combustion due to the generation of anthracene, fluoranthenes and pyrene [57]. This could be mitigated with the installation of Urea-based SCR systems, LNT Lean NOx Trap, or Exhaust Gas Recirculation that reduces the combustion temperature [102]. Finally, freshwater eutrophication should be reduced by 61% for these biofuels. Although one could think this is the consequence of the water used for growing the algae, cultivation is typically carried out in closed circuits where water is recirculated. Consequently, 90% of the freshwater eutrophication stems from the use of energy from the grid, which, in the case of the US-WECC power mix, is dominated by coal (34%) and natural gas (18%).

Therefore, trying to meet this target implies either generating the required energy internally using cleaner sources or relying on a more sustainable energy mix. This latter option is explored in more detail in the next section.

3.3. Enhancement scenarios

After identifying hotspots for inefficient biofuels in the previous section, we next quantify the impact of adopting certain supply and demand-side measures to improve their sustainability performance. For the former, we focus on an improvement measure recurrently identified as promising in the previous section, considering only nominal values for the indicators, namely the use of a renewable-based electricity matrix to supply energy for foreground processes. The mix proposed follows the guidelines of the European Green Deal [12] for reduction of GWP emissions and is based on 74% hydroelectric, 25% geothermal and 1% wind. To complement the demand-side measures, we also analyze the significance of adopting different demand-side measures, represented here by the use of different vehicles and passenger loads. Four scenarios are considered in this regard on top of the reference case discussed so far (i.e., labelled as scenario LVO1): light vehicles at minimum capacity (i.e., one passenger, LV1); light vehicles at maximum capacity (i.e., five

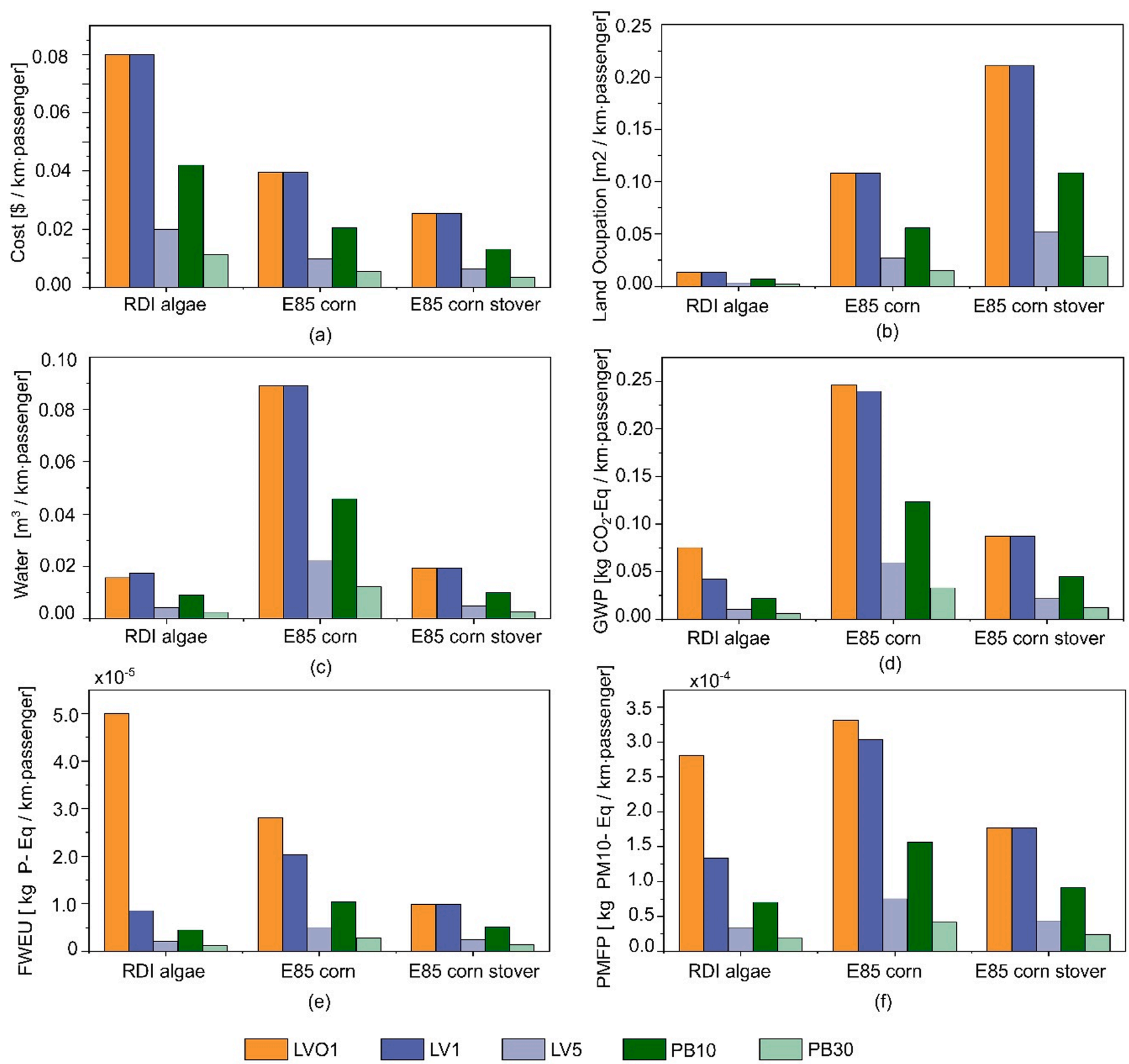

Fig. 8. Changes achieved in inputs (i.e., costs, land occupation and water use) and selected undesirable outputs (i.e., global warming potential, freshwater eutrophication and fine particulate matter formation) by replacing the current mix (i.e., US-WECC) and vehicle type/usage for three fuels obtained from different types of carbon sources: bio-oil, cellulose and first-generation feedstock. LVO1: base case without modifications (1 passenger); LV1: modified base case (1 passenger); LV5: modified base case (5 passengers); PB10: public transport (10 passengers); PB30: public transport (30 passengers).

passengers, LV5) and the use of public transport (i.e., a bus) at 30% of its maximum capacity (i.e., ten passengers, PB10) and at maximum capacity (i.e., 30 passengers, scenario PB30). All these scenarios adopt the sustainable mix.

The consequences of adopting such measures and scenarios are calculated for a subset of six of the 12 performance indicators considered so far (i.e., cost, land occupation, water use, global warming potential, freshwater eutrophication and fine particulate matter formation) for three different biofuels (i.e., RDI based on algae, E85 based on corn and E85 based on corn stover). The results obtained are shown on a per-capita basis in Fig. 8, where a comparison of scenarios LV1 and LVO1 allows us to assess the impact of the *ceteris paribus* change of the electricity source.

Modifying the electricity matrix (i.e., comparison of scenario LV1 with LVO1) allows algae-based RDI to achieve important reductions in FWEU (82%), PMFP (52%) and GWP (44%). This change can be explained by the high electricity requirements of algae-based biofuel production and would suffice to attain the targets suggested by DEA for some inefficient biofuels such as algae BD20 (e.g., 61% FWEU, 18% PMFP). In contrast, improvements in these three indicators for E85 biofuels are inexistent. This is because their production process requires thermal energy rather than electricity. Although not explored here, the implementation of heat pumps for waste heat recovery [103] could help these fuels to meet their improvement targets. Similarly, the remaining three indicators (i.e., cost, land occupation and water use) are barely affected by the change of the electricity mix, which suggests that other measures would need to be pursued before improvement targets can be attained. For the case of cost reduction, governments can play a key role by providing economic incentives for biofuel production or discounting certain taxes for the production or sale of biofuels. This would help alleviate the economic burden of some alternatives that can be key in the achievement of environmental targets, that are becoming more demanding. Meanwhile, we note that adopting the latest standards in farming practices is not the only way to pursue improvements in land occupation and water use: increasing the efficiency of processes downstream the supply chain (i.e., hydrolysis of lignocellulosic materials [104] or the fermentation process [105] will ultimately result in a lower demand for biomass feedstock and, therefore, lesser impacts from farming.

More optimistic improvements are observed in all the cases when supply and demand-side measures are combined (i.e., comparison of scenario PB30 with LVO1). Revisiting the case of algae-based RDI, reductions in FWEU, PMFP and GWP reach values as high as 97%, 93% and 92%; on average, 68% higher than in scenario LV1. Similar patterns are also observed for the rest of the biofuels and indicators, which, in this case, achieve improvements between 86% and 90%. These would allow meeting the improvement targets requested by DEA for all the indicators in the case of E85 from corn stover and almost all the indicators except for land occupation, water use and terrestrial ecotoxicity in the case of corn-based E85.

These results highlight the importance of demand-side measures, often overlooked in biofuel studies [106,107], since adopting a responsible behavior can be, at least, as impactful as shifting to cleaner energy sources. Indeed, the greater the number of passengers in a certain vehicle, the greater the improvement in the performance of biofuels. The only exception to this rule is the use of public transport at 30% of its capacity, which results in a worse alternative than a light vehicle with five passengers.

4. Conclusion

In an effort to identify patterns that can aid in the development of effective policies ensuring the sustainability transition in the transportation sector, we combined LCA with DEA to assess the performance of 72 biofuel routes through the lens of sustainability. The different alternatives result from the combination of 19 biological feedstocks, four biofuel production processes and five biofuel blends.

The biofuel alternative with the highest efficiency score was based on MSW, which suggests that these should be prioritized over other carbon sources. Fuels from natural oils also show a promising performance, with 20 of the 22 units analyzed deemed efficient. Among the remaining carbon sources, results agree with the recent trend of promoting the use of cellulosic material for ethanol production. In terms of fuel type, our results suggest that policies should favor the widespread adoption of renewable diesel over traditional ethanol or biodiesel, since the former achieved the best performance thanks to a higher fuel economy and a higher biogenic carbon content in the fuel. The fuel type, however, was not found as impactful as the carbon source in achieving high efficiency scores.

To complement policies for regulating biofuel supply, we also explored the effectiveness of demand-side measures for the transportation sector. We found that adopting responsible practices for vehicle use could bring even more benefits than improving the biofuel production processes or using cleaner energy sources.

Finally, our analysis also provided targets for the improvement of inefficient biofuels that, if attained, would make them efficient. In this regard, reductions in land occupation and water use, although highly relevant and often identified as key in our results, might not be possible to achieve depending on the type of crop and region. A case-by-case analysis is necessary for the farming stage to avoid the transportation of biomass over long distances and ensure no risks are imposed on food security. Indeed, the most appropriate feedstock might depend on the region of interest.

Promoting biological carbon sources today as an interim solution for the transportation sector might proof useful even if the future is finally dominated by electric vehicles since the infrastructure created for growing and transporting the biomass today could still be exploited by bioenergy plants tomorrow. If combined with carbon capture and storage, these plants will remove carbon dioxide from the atmosphere, a strategy deemed essential for meeting net-zero targets. In this context, multi-criteria approaches, such as the one presented in this contribution, offer a powerful framework to perform holistic assessments with the capacity to minimize burden-shifting episodes and aid policy-makers in the development of better-informed policies.

CRediT authorship contribution statement

Richard Cabrera-Jiménez: Conceptualization, Methodology, Software, Writing – original draft, Visualization. **Josep M. Mateo-Sanz:** Methodology, Writing – review & editing. **Jordi Gavaldà:** Validation, Writing – review & editing. **Laureano Jiménez:** Resources, Writing – review & editing, Supervision, Project administration. **Carlos Pozo:** Conceptualization, Methodology, Validation, Writing – review & editing, Supervision.

Declaration of Competing Interest

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

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apenergy.2021.118201.

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