



DRIVING TOWARDS SUSTAINABILITY: DECISION SUPPORT TOOLS TO GUIDE THE ENERGY TRANSITION IN THE TRANSPORT SECTOR

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**UNIVERSITAT
ROVIRA I VIRGILI**

**Driving towards sustainability: Decision
support tools to guide the energy
transition in the transport sector.**

RICHARD EMMANUEL CABRERA JIMÉNEZ

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Driving towards sustainability:
Decision support tools to guide
the energy transition in the
transport sector

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I state that the present study, entitled “Driving towards sustainability: Decision support tools to guide the energy transition in the transport Sector.”, presented by Richard Emmanuel Cabrera Jiménez for the award of the degree of Doctor, has been carried out under our supervision at the Department of Chemical Engineering of this university.

Tarragona, 28th November 2023

Doctoral Thesis Supervisor.

Dr. Carlos Pozo Fernandez

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Summary

The imperative shift toward a low-carbon future requires a profound evaluation of sustainable alternatives in almost all aspects of our lives. This thesis focuses on biofuels and electric transport technologies, aiming to provide a comprehensive understanding of their short, medium, and long-term impacts on the environment and society. Recognizing the persistent dependence on liquid fuels, especially in heavy transport and developing regions, biofuels are positioned as a crucial transitional step until widespread electric vehicle adoption.

This thesis endeavors to address emerging challenges within the transportation sector and facilitate a sustainable energy transition by providing guidance for effective sustainability policies. Despite the substantial challenges associated with the use of biofuels, the thesis recognizes their significant potential for environmental improvement compared to current fuels. In response to this recognition, the study emphasizes the importance of informed decision-making among various stakeholders, including research centers, companies, and policymakers. To navigate the intricate landscape surrounding biofuels, the thesis addresses three specific challenges related to biofuel adoption, offering practical examples that demonstrate the effective utilization of specific tools for informed decision-making.

Firstly, it addresses the selection of optimal biofuel alternatives for light-duty vehicles. Multicriteria Decision-Making (MCDM) tools, particularly Data Envelopment Analysis, are employed, guided by a comprehensive Life Cycle Assessment (LCA) approach.

The second area of focus delves into the optimal design of a biofuel production process, with a specific focus on the emerging technology of microalgae-based biofuels for heavy transport. In this stage, simulation tools are employed to understand factors such as raw material use, energy consumption, and emissions. This analysis is further enriched by incorporating an Absolute Environmental Sustainability Assessment (AESA) method, utilizing the Planetary Boundary (PB) framework.

The third area involves formulating environmental policies to maximize the reduction of greenhouse gas emissions within the transport sector, accounting for anticipated technological advancements. The thesis centers on road freight transport, examining the viability of biofuel-powered trucks and electric trucks as alternatives to diesel trucks. Through *ex-ante* LCA techniques, the study provides a comprehensive understanding of the global impact of these transportation alternatives, contributing valuable insights to the ongoing discourse on sustainable transport.

In conclusion, the thesis advocates prioritizing renewable diesel over traditional ethanol or biodiesel due to its superior performance, emphasizing its importance in achieving decarbonization goals. The exploration of emerging technology such as microalgae biofuels reveals promising routes for climate change mitigation and reduced biosphere impacts. Additionally, the comprehensive assessment, considering the medium and long-term evolution of technologies, identifies viable alternatives for the decarbonization of freight road transport until 2100, with Battery Electric Trucks emerging as a promising long-term option. Results also emphasize the continued importance of biofuels, in the short to medium term.

Resumen

El cambio necesario hacia un futuro con bajas emisiones de carbono exige una profunda evaluación de las alternativas sostenibles en casi todos los aspectos de nuestras vidas. Esta tesis se centra en los biocombustibles y las tecnologías de transporte eléctrico, con el objetivo de proporcionar una visión global de sus repercusiones a corto, medio y largo plazo sobre el medio ambiente y la sociedad. Dada la elevada dependencia de los combustibles líquidos, especialmente en el transporte pesado y en las regiones en desarrollo, los biocombustibles se posicionan como un paso de transición crucial hasta la adopción generalizada del vehículo eléctrico.

Esta tesis pretende abordar los retos emergentes en el sector del transporte y facilitar una transición energética sostenible proporcionando pautas para la elaboración de políticas de sostenibilidad más eficaces. A pesar de los importantes retos asociados al uso de biocombustibles, la tesis cuantifica su notable potencial de mejora medioambiental en comparación con los combustibles usados actualmente. En respuesta a este hecho, el estudio hace hincapié en la importancia de tomar decisiones basadas en evidencias entre los diferentes actores involucrados, incluidos los centros de investigación, las empresas y los responsables políticos. Para poder afrontar el intrincado panorama que rodea a los biocombustibles, la tesis aborda tres retos específicos relacionados con la adopción de biocombustibles, ofreciendo ejemplos que demuestran la utilización eficaz de herramientas específicas para la toma de decisiones .

En primer lugar, se aborda la selección de alternativas óptimas de biocombustibles para vehículos ligeros. Se emplean herramientas de toma de decisiones multicriterio (MCDM), en particular el Análisis Envolvente de Datos (DEA), guiadas por un enfoque integral de Análisis del Ciclo de Vida (ACV).

La segunda área de interés profundiza en el diseño óptimo de un proceso de producción de biocombustibles, con especial atención a

la tecnología emergente de biocombustibles basados en microalgas destinados al transporte pesado. En esta etapa, se emplean herramientas de simulación para comprender factores como la selección de materias primas, el consumo de energía y las emisiones generadas. Este análisis se enriquece aún más incorporando un método de evaluación absoluta de la sostenibilidad medioambiental (AESAs), utilizando el marco del Límite Planetario (PB).

La tercera área consiste en formular políticas medioambientales para maximizar la reducción de las emisiones de gases de efecto invernadero en el sector del transporte, teniendo en cuenta los avances tecnológicos previstos. La tesis se centra en el transporte de mercancías por carretera, examinando la viabilidad de los camiones alimentados por biocombustibles y los camiones eléctricos como alternativas a los camiones diésel. Mediante técnicas de ACV *ex ante*, el estudio proporciona una comprensión exhaustiva del impacto global de estas alternativas de transporte, aportando valiosas ideas al actual debate sobre el transporte sostenible.

En conclusión, la tesis recomienda priorizar el diésel renovable sobre el etanol o el biodiésel tradicionales debido a su rendimiento superior, destacando su importancia para alcanzar los objetivos de descarbonización. La exploración de tecnologías emergentes como los biocombustibles de microalgas revela rutas de producción que son prometedoras para la mitigación del cambio climático y la reducción de los impactos sobre la biosfera. Además, la evaluación exhaustiva, teniendo en cuenta la evolución de las tecnologías a medio y largo plazo, identifica alternativas viables para la descarbonización del transporte de mercancías por carretera hasta 2100, con los camiones eléctricos usando baterías como alternativa emergente a largo plazo. Asimismo, el estudio subraya la importancia de los biocombustibles, a corto y medio plazo.

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CHAPTER I.

Introduction

I. Introduction

1.1 Background and motivation

The international community is increasingly focused on addressing the challenges of climate change and its detrimental impacts associated with rising carbon emissions. The transition to a low-carbon growth agenda forms the heart of contemporary economic development debates.[1–3]

The transportation sector, identified by the International Energy Agency as the third-largest global carbon emitter [4], demands urgent attention due to its important impact on global warming. Numerous studies underscore the challenges associated with achieving energy savings and emissions reductions in this sector [5]. In response, the need for low-carbon fuel alternatives becomes clear. Among these alternatives, biofuels, synthetic fuels, and the adoption of electric vehicles (EVs), stand out and are considered in most climate change policies.

Despite the optimism surrounding intensive scenarios for EV sales, studies suggest that, at best, EVs might constitute 50% of the overall vehicle fleet by 2050. This projection is grounded in the slow turnover rates of existing vehicles. In this sense, even with 100% of newly sold vehicles being electric, a complete transition to an all-electric fleet would take between 20 to 25 years from today [6].

As a result, the transport sector is anticipated to maintain its dependence on liquid fuels, at least in the medium term. This reliance is particularly crucial in sectors such as heavy transport and developing countries, where limitations in infrastructure investment could impede the swift adoption of EVs [6]. Indeed, policies actively endorse the short-term use of biofuels as an immediate alternative, serving as a transitional step toward the widespread adoption of EVs.

Currently, the predominant liquid fuel alternative involves biofuels, commercially produced from crops also utilized for food and feed, named as first-generation fuels (e.g., corn, soybeans, sugarcane) [7]. This reliance on

biofuels derived from food crops has sparked considerable opposition to their use due to concerns about diverting resources from food production and the resultant increase in food prices. Nonetheless, a wealth of potential lies in the production of biofuels sourced from non-food crops, dedicated energy crops, such as crop and forestry residues (i.e., second generation biofuels) or microalgae (i.e., third generation biofuels) [8]. This shift to sustainable sources presents an opportunity to overcome the drawbacks associated with first-generation biofuels.

The close relationship and dependence on current biofuels raises the question of land availability to significantly scale up biofuel production. Since both first and second-generation biofuel feedstocks inevitably compete for land and water use, and other factors must also be undertaken. For this reason, the exploration of new alternatives for the transportation sector involves, not only addressing the GWP, but also scrutinizing other environmental indicators such as water contamination, human health or toxicity, ensuring a holistic assessment of biofuel sustainability.

As highlighted earlier, the utilization of biofuels poses significant challenges. However, it also gives an alternative with considerable potential for environmental improvement compared to current fuels. In light of this, various stakeholders, including research centers, companies, and policymakers, must make informed decisions to channel efforts and resources towards specific biofuel alternatives. To navigate this complex landscape, they require the support of various tools. Hence, this thesis addresses three specific challenges related to biofuels offering practical examples of how to leverage specific tools for informed decision-making.

The first challenge centers around selecting the optimal biofuel alternatives for light-duty vehicles among those currently available (see section 2). To achieve this, the analysis begins with a comprehensive review of literature data spanning from feedstock production to vehicle operation. Multicriteria decision making (MCDM) tools will be employed for the selection process, facilitating the integration of environmental, social, and economic metrics derived from the literature. The environmental impact assessment will utilize the Life Cycle Assessment (LCA) approach.

The chosen MCDM tool is Data Envelopment Analysis (DEA), DEA facilitates the identification and selection of the best alternatives. Moreover, it enables the pinpointing of essential parameters in alternatives that exhibit potential for improvement. This identification process allows us to develop specific technologies guided by insights into what improvements should be made in certain alternatives to become efficient. DEA model will be further discussed in section 2.2.

In addressing the second challenge, we delve into the optimal design of a biofuel production process, focusing on an emerging technology—microalgae-based biofuels for heavy transport. Employing simulation tools provides a comprehensive understanding of factors like raw material procurement, energy consumption, and emissions.

To further enrich our evaluation, we incorporate an Absolute Environmental Sustainability Assessment (AESAs) method. Within AESAs, the Planetary Boundary (PB) framework serves as a metric to quantify the state of the transportation sector under biofuel use. This framework offers insights into potential effects within defined limits, aligning with the ecological capacity of the planet. This goes beyond conventional Life Cycle Assessment (LCA), which only permits comparative assessments. Section 3.2. will delve into the detailed development of the PB framework for heavy transport.

Finally, the third challenge addressed involves formulating environmental policies to reduce greenhouse gas emissions as much as possible. These proposed policies, designed for both the medium and long term, take into account anticipated technological advancements (see section 4.2). Addressing the challenge requires the incorporation of an evaluation method capable of considering technological changes over time in a specific sector, aiding policy-makers in developing robust environmental strategies.

In this thesis (in section 4), we focus on road freight transport, specifically studying the viability of biofuel-powered trucks (bioICT) and electric trucks (BET) as alternatives to diesel trucks. To achieve this, we employ ex-ante LCA techniques, offering a comprehensive understanding of the global impact of these transportation alternatives. Through this comparative analysis, we aim to shed light on the potential environmental and human impacts of these

promising alternatives, contributing to informed regulations that align with current CO2 emission reduction targets for the transportation sector.

Figure I-1- summarizes the methods applied in this thesis, summarizing the three challenges and the tools used to solve them.

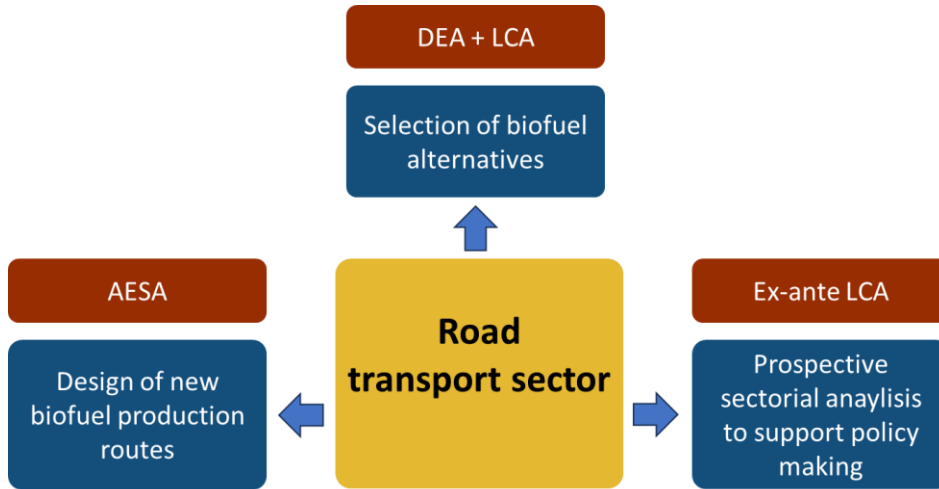


Figure I- 1 Overview of the methods performed in the thesis.

1.2 General objectives

The primary goal of this thesis is to address emerging challenges in the transportation sector and the energy transition by guiding its sustainability policies. It aims to achieve this by integrating decision-making support tools based on mathematical programming (i.e., optimization) and the LCA methodology. These tools will help to identify efficient processes and assess the potential environmental impacts throughout the transport activities life cycle.

To achieve the overall goal, the following specific objectives must be met:

- Find a methodological approach for comparing existing biofuel production alternatives, integrating diverse sustainability metrics to identify the most favorable options .
- Conduct an environmental impact assessment of biofuel production, emphasizing a holistic approach that considers both comparative

assessments between fossil fuel alternatives and compliance with the planet's ecological capacity.

- Propose policies aimed at maximizing the reduction of GHG emissions within the transport sector. This includes accounting for the medium and long-term evolution of technologies in the sector, considering their multi-regional effects on a global scale.

1.3 Multicriteria decision tools

The term multicriteria decision-making describe a collection of formal approaches based on mathematical tools used to structure and solve decision and planning problems containing multiple conflicting criteria and numerous alternatives.

Evaluating and ranking a set of alternatives in real-world applications is a challenging task for many decision-makers, particularly when the number of alternatives and criteria increases. The use of such tools for addressing energy planning has been extensively researched, including environmental, socio-economic, and technical barriers. [9]

1.3.1 Data envelopment analysis

Data Envelopment Analysis (DEA) is a mathematical programming technique first introduced in 1978 by Charnes, Cooper, and Rhodes [10], with the aim of comparing and evaluating a homogeneous set of decision-making units (DMUs) in a production system with multiple inputs and multiple outputs.

Subsequent developments have proved DEA as a valuable tool for performance evaluation in a wide number of fields, with interesting applications in health care, banking, manufacturing, and energy planning. In the latter field, DEA has proven its usefulness in solving environmental, socio-economic, and technical barriers inherent to this sector. [11]

DEA stands out from other multi-criteria assessment methods due to its ability to combine various indicators into one performance score, without the need to establish subjective weights between the indicators. This is particularly useful in sustainability assessments that always crates controversy. Also, it enables the integration of indicators covering all three sustainability dimensions into a single metric, allowing for an easy identification of efficient and inefficient alternatives (see section 2.2.).

Furthermore, DEA can provide information on the extent to which inefficient alternatives should improve to become comparable with the top-performing processes.

DEA is a non-parametric LP technique that analyses a set of comparable DMUs individually by solving a set of LP model, identifying those that exhibit the best performance. DEA returns a performance score, also called efficiency score, lying between 0 and 1. DMUs with a score of 1 are referred to as efficient and are linearly combined to form an efficient frontier, which is a linear function connecting all the efficient DMUs. Meanwhile, DMUs with a score strictly lower than one 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.

The original input-oriented DEA CCR model (after its creator's names Charnes-Cooper-Rhodes) was first proposed by Charnes et al. [10]. It is a nonlinear model that measures the efficiency of a DMU as the ratio of the weighted sum of their outputs and to the weighted sum of their inputs. This model aims to find the optimal weights so that the efficiency of the DMU being analyzed is maximized with respect to the remaining DMUs.

Mathematically, the DEA CCR model considers a set of j DMUs ($j=1,\dots,n$), each of them using i inputs x_{ij} ($i=1,\dots,m$) to produce r outputs y_{rj} ($r=1,\dots,k$), and can be formally posed as follows (Eq. 1-1):

$$\begin{aligned} \max \quad & \theta_{j_0} = \frac{\sum_{r=1}^k u_r y_{rj}}{\sum_{i=1}^m v_i x_{ij}} && \text{Eq. (1-1)} \\ \text{s. t.} \quad & \sum_{r=1}^k u_r y_{rj} - \sum_{i=1}^m v_i x_{ij} \leq 0 \quad j = 1, 2, \dots, n \\ & u_r, v_i \geq 0 \quad r = 1, 2, \dots, k ; i = 1, 2, \dots, m \end{aligned}$$

where θ_j represents the efficiency score of the DMU_j and u_r and v_i are corresponding output and input weights chosen for DMU_j. If DMU_j satisfies that $\theta_j = 1$, it is deemed efficient; yet when $\theta_j < 1$ it is considered inefficient.

As previously mentioned, the original input-oriented CCR DEA model [10](Eq. I-1) is both nonlinear and nonconvex. However, it can be converted into the following (linear programming) LP model (Eq. I-2), where the denominator is set to one and the numerator is maximized:

$$\begin{aligned}
 \max \quad & \theta_{j_o} = \sum_{r=1}^k u_r y_{rj_o} \\
 \text{s. t.} \quad & \sum_{i=1}^m v_i x_{ij_o} \leq 0 \quad i = 1, 2, \dots, m \\
 & \sum_{r=1}^k u_r y_{rj} - \sum_{i=1}^m v_i x_{ij} \leq 0 \quad j = 1, 2, \dots, n \\
 & u_r, v_i \geq 0 \quad r = 1, 2, \dots, k ; i = 1, 2, \dots, m
 \end{aligned} \tag{Eq. (I-2)}$$

where the subscript j_o denotes the specific DMU under assessment.

For this primal LP problem, a dual partner problem (duality) can provide the same information as the primal model (Eq. I-2), i.e., efficiency scores, while also calculating targets for inefficient DMUs to enhance efficiency. The LP DEA dual model (Eq. I-3) is formulated by assigning one dual variable to each constraint in the primal model as follows [12]:

$$\begin{aligned}
 \min \quad & Z = \theta_o - \varepsilon \left(\sum_{r=1}^k S_r^+ \sum_{i=1}^m S_i^- \right) \\
 \text{s. t.} \quad & \sum_{j=1}^n \lambda_j x_{ij} + S_i^- = \theta_o x_{i_o} \quad i = 1, 2, \dots, m \\
 & \sum_{j=1}^n \lambda_j y_{ij} - S_r^+ = y_{r_o} \quad r = 1, 2, \dots, n
 \end{aligned} \tag{Eq. I-3}$$

$$\lambda_j, S_i^-, S_r^+ \geq 0 \quad r = 1, 2, \dots, k; i = 1, 2, \dots, m; j = 1, 2, \dots, n$$

Where ε is a non-Archimedean value to ensure that efficiency always has a positive value, the efficiency of the DMU' under consideration is measured by an unconstrained θ_o , which is always less than or equal to 1 ($\theta_o \leq 1$). S_r^+ and S_i^- are slack variables that represent the additional amount by which an input (or output) should be reduced (or increased) to become strongly efficient, more details in section 2.2.3. Note that the values of the slacks are all zero (S_r^+ and $S_i^- = 0$) in the efficient units ($\theta_o = 1$), and strictly positive in the inefficient ones ($\theta_o < 1$). λ_j is a variable that represents the weight assigned to each efficient DMU (belonging to the efficient frontier). These weights are used to form a virtual, non-existing efficient unit that can be used as a benchmark to improve a particular inefficient unit.

This hypothetical virtual unit is obtained by projecting the inefficient unit radially on the efficient frontier. The BCC dual model can be formulated by adding a convexity constraint to Eq. I-3 which ensures $\sum_{j=1}^n \lambda_j = 1$ [13]

To illustrate DEA concepts, we present Fig. I-2, with an application of the CCR and BBC DEA models; these examples present a set of six DMUs (A to F). Under the CCR model assuming constant return to scale, the efficient frontier is given by the line starting from the origin and passing through the efficient units C and E. In this model, only DMUs C and E are efficient since none of the others can produce a higher output to input ratio. In the case of the BCC model, DMUs A and F continue to be efficient, and now DMUs A and F become efficient as well, to form what is called the reference set, due to the variable returns to scale property of this model. The frontier composed by the efficient DMUs is called strongly efficient frontier. On the other hand, two more segments are also shown as part of the efficient frontier, which are represented by gray dotted lines. These segments are formed by the distant efficient DMUs and go to infinity parallel to the output and input axes; they are called weakly efficient frontier.

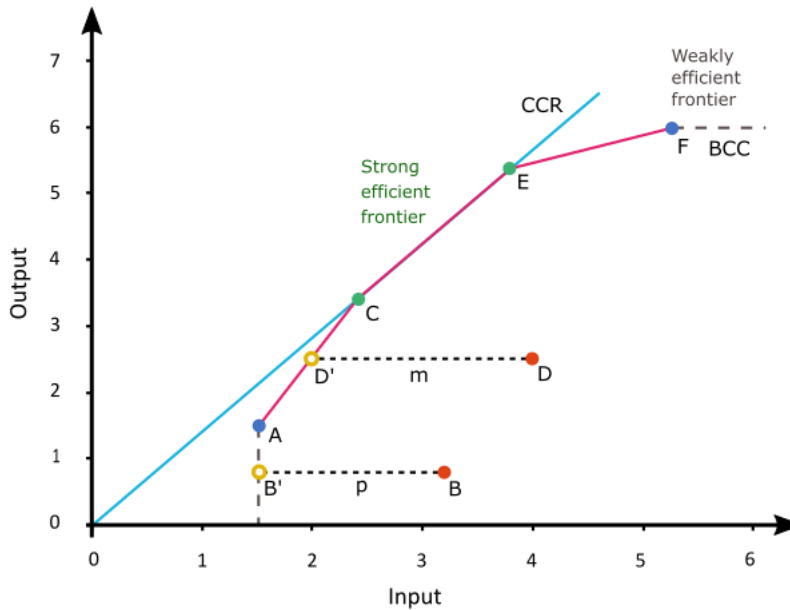


Figure I- 2. Example of CCR and BCC DEA models

On the other hand, DMUs B and D (presented in red) are considered inefficient regardless of the model used. To obtain the improvement targets of these inefficient DMUs, they are projected in the efficient frontier, thus forming their corresponding virtual DMUs (i.e., B' and D').

The projections used by the BCC model to project the virtual DMUs onto the efficient boundary are radial, being output oriented (horizontal) or input oriented (vertical).

The previous DEA CCR model (Eq. I-2) is called input-oriented; this means that inefficient units are turned efficient by reducing the inputs proportionally while maintaining the same level of outputs. On the other hand, it is possible to reformulate the equations to obtain an output-oriented model. In this case, an inefficient unit would become efficient by increasing its outputs while keeping the inputs constant. Hence, the model's orientation determines how inefficient DMUs are projected onto the efficient frontier, additional details on this topic will be provided in section 2.2.

Furthermore, the CCR model undertakes constant returns to scale (CRS), assuming that DMUs operate at the same scale, where outputs change proportionally with the change in inputs. Alternatively, the DEA model allows for a formulation that

considers variable returns to scale (VRS), commonly known as the BCC DEA model (after its creator's names Banker, Chames and Cooper[14]. The selection of model orientation and the returns-to-scale type depends on the specific application under consideration [15].

However, it is also possible to make other types of projections, as is the case of the slack based measurement (SBM) model, which is discussed and used in section II of this thesis.

For further information about DEA models and extensions, the reader is referred to Cook and Seiford [13], Cooper et al. [12] and Tone et al. [16].

1.3.2 Super-efficiency model

In order to further discriminate the performance of efficient DMUs, Andersen and Petersen [17] proposed the concept of super efficiency in DEA. Initially, the super-efficiency model involved standard DEA models (CRS or VRS) that rank efficient DMUs assigning to each of them efficiency scores beyond one. This is obtained by assuming that the DMU to be evaluated is excluded from the reference set. An additional usefulness of this super-efficiency model is that it can also be considered a measure of stability for efficient DMUs. (see section 2.2.4)

Figure I-3 shows an example of the super-efficiency classification using four efficient DMUs. In this super efficiency model, the efficiency of each DMU is evaluated against a new efficient frontier that is formed when the DMU studied is excluded from the frontier.

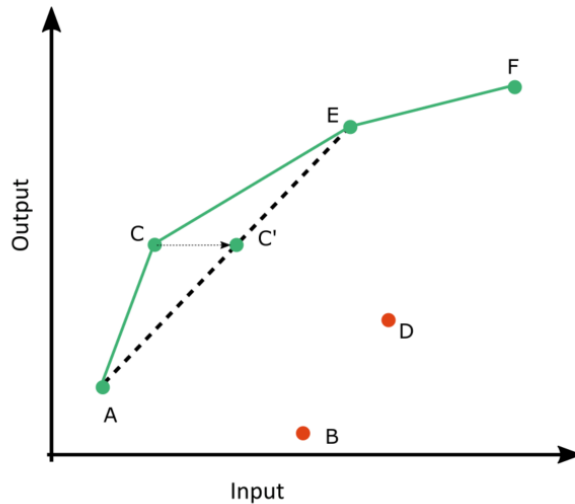


Figure I- 3. Example of super-efficiency method

If we analyze the DMU C, the efficient frontier resulting from this modification will be given by the line \overline{AEF} . When projecting C to the new efficient frontier, it becomes point C', which is still efficient as it is part of the (new) efficient frontier, yet it has additional inputs compared to the original DMU. Hence, the original DMU is considered super-efficient, as it performs even better than an efficient DMU.

Another typical group of super-efficiency measures are developed by dealing directly with input and output slacks. These non-radial measures include SBM super-efficiency [18] and additive super-efficiency [19]. The SBM super-efficiency will be further discuss in section II subsection 2.2.4.

1.4 Environmental impact assessment

This thesis addresses the sustainability challenge of transport sector by evaluating the environmental impact of different alternatives for transport. Among the wide range of environmental impact assessment methodologies, Life Cycle Assessment (LCA) has become the predominant methodology, being broadly applied both in private and public environmental decision-making contexts [11,20–22]

So far, several impact assessment methods have been put forward (e.g., CML, ReCiPe), yet they lack absolute thresholds to elucidate whether a given system should be deemed sustainable. To overcome this limitation, we resort to an

absolute environmental sustainability assessment (AESAs) method, which bridges conventional LCA principles with the concept of Planetary boundaries developed by Rockstrom et al. [23] and Steffen et al.[24]

1.4.1 Life cycle assessment

LCA is a method that enables the quantitative evaluation of the environmental impact of goods and services throughout their entire life cycle. An LCA study encompasses four phases: i) defining the goal and scope, ii) conducting an inventory analysis, iii) performing the impact assessment, and iv) interpreting the results.

The four phases of a LCA study process are described next into detail:

The first phase of an LCA study consists of define the goals and scope in order to guide the entire process. The goal and spatial and temporal scope of the study are set, as well as the functional unit and system boundaries. It is also important to choose between attributional or consequential modelling approaches, as this decision will affect the next step (i.e., life cycle inventory), as well as system boundaries and other methodological choices. [25]

The attributional approach is currently the most commonly used method in LCA to assign responsibility for a set of impacts. In this method the main aim is to assess the environmental impacts of a product/service assuming that the system under study is not able to change the available production capacity of the market. On the other hand, in the consequential LCA, the aim is to describe how environmentally relevant flows will change in response to possible decisions [26].

In this phase, the method used to divide environmental burdens among different products from the same process is also selected. Two main options are available: system expansion and allocation. System expansion considers products and co-products as different alternatives to assign environmental burdens, being recommended by the ISO [27]. However, when it is not possible to use system expansion, the allocation approach can be applied by apportioning the impacts according to weights (such as mass, economic, or energy allocation), further details are explained in section 2.2.

The second phase of the LCA process is the life cycle inventory analysis (LCI), where all inputs and outputs of the system are collected (i.e., emissions and resource usage), measured, or estimated, and tracked throughout the life cycle.

This stage usually requires an iterative process with the scope and goal definition to adjust boundaries and allocations. The overall system is broken down into small units where all inputs and outputs are collected from different sources. At this stage, a large amount of data must be obtained, which eventually generates uncertainty. To streamline this process, several commercial software and open-source models have been developed, with options such as SimaPro [28], Gabi [29], openLCA [30] and Brightway2 [31]. However, the effective utilization of these tools requires the availability of what is called background data, i.e., all activities not explicitly modelled including energy and materials that are delivered to the foreground system, more details are discussed in sections 2.2.1, 3.2.1 and 4.2. To address this requirement, various LCA databases are accessible, with Ecoinvent standing out as a leading and comprehensive resource [32].

The process of the inventory can be mathematically expressed as follows:

$$LCI_i^{TOT} = \sum_j LCI_{i,j} \quad \forall i \quad (\text{Eq. I-4})$$

where LCI_i^{TOT} represent the total amount of a specific flow i (e.g., carbon dioxide) which is computed as the summation of all the flows i for all the system units j , $LCI_{i,j}$.

The third phase consists of the life cycle impact assessment (LCIA). During the LCIA stage, all the gathered data on emissions and resource usage is converted into impact categories and analyzed for environmental impacts using characterization factors. LCIA methodologies vary regarding the impact categories covered, impact indicators, and geographical focus.

Once the characterization process is complete, the next optional steps are normalization and weighting. Normalization ensures that all impact indicators are in a common metric, making them comparable, while weighting groups of different impact categories based on agreed subjective valuations in the scientific community [27]. The impact characterization process can be summarized as follows:

First, the life cycle inventory (LCI) data is transformed into impact referring to the midpoint categories using Eq. I-6.

$$I_m = \sum_i C_{mi} \cdot LCI_i^{TOT} \quad \forall m \quad (\text{Eq. I-6})$$

In the equation I-6, C_{mi} denotes the characterization factor that connects the flow i with the midpoint impact category m . As a result, I_m represents the indicator for the midpoint category m . These midpoint category indicators can then be aggregated into endpoint damage categories, as stated in Eq. I-7:

$$I_e = \sum_m N_{me} \cdot I_m \quad \forall e \quad (\text{Eq. I-7})$$

Where N_{me} denotes the normalization factors that relate the midpoint categories m with the endpoint category e , allowing aggregation into the endpoint categories e (I_e). Finally, endpoint category indicators can be further aggregated into a single final score (S) using equation Eq. I-8

$$S = \sum_e W_e \cdot I_e \quad (\text{Eq. I-8})$$

Where the endpoint indicators I_e are multiplied by weighting factors W_e , the impact scores can be converted into the same units (e.g., points) and then summed to produce the weighted impact score S .

The last phase of the LCA study is devoted to results interpretation. This involves drawing conclusions from the study to provide recommendations for the decision-makers. While the results are useful in identifying areas where efforts should be made to minimize the environmental impact, they do not provide clear guidelines or targets for achieving optimal reduction. However, it is important to note that the results alone are not enough to provide clear guidelines or targets for achieving optimal reduction. Environmental decision-making problems can be complex, involving the comparison of several alternatives and considering various objectives across different scenarios. Additionally, since these problems are interdisciplinary, there are usually many stakeholders involved, which makes decision-making even more challenging. To overcome these challenges, specialized tools and techniques may be required to facilitate decision-making.

After quantifying the burdens and impacts throughout the entire life cycle, either by following the previous four phases or by collecting data from an LCA database, mathematical programming models can be used to incorporate LCA outcomes (see

section 2.2.). This enables the identification of the best solutions according to environmental principles in a systematic manner. By integrating LCA's inputs and impact data into mathematical models, mathematical programming can systematically identify the best alternatives in terms of environmental, technical, economic, or social criteria. This capability has been exploited by authors combining tailored approaches to guide practitioners and improve environmental performance, providing insights for decision-makers and policymakers through strengthened complementarities. [15,33,34]

As previously mentioned, conducting a LCA enables the quantification of the environmental impacts associated with a given product, which are subsequently integrated as parameters into mathematical programming models addressing the problem or specific product, further details are explained in section 2.2 and section 3.2.

1.4.2 Absolute environmental sustainability assessment

LCA enabled the performance of extensive environmental analyses of processes and products, encompassing various activities ranging from feedstock extraction to waste management.

So far, several assessment methods have been proposed based on anthropogenic alterations to the global environment resulting in climate change, biodiversity loss or pollution (e.g., CML and ReCiPe), which often approached as separate issues, ignoring their nonlinear interactions and their aggregate effects on the overall state of the Earth system [23]. Moreover, there are no absolute thresholds for determining whether a given system should be considered sustainable.

To overcome this limitation, here we focus on an absolute environmental sustainability assessment (AESAs) method, which bridges conventional LCA principles with the concept of Planetary boundaries (PBs), developed by Rockström et al., and later updated by Steffen et al., aiming at quantifying the absolute environmental sustainability level of human activities. [23,24]

The PB framework aims to provide a scientific understanding of the impact of human activities on the global environment. It considers the state of the Earth system as a whole and proposes nine biochemical and bio-geophysical boundaries. These boundaries define a "safe operating space for humanity" (SOS) for each Earth system that regulates the state of the planet within ranges that have been

historically known and scientifically proven to maintain Earth's stability. This stability will ensure life-support systems that are appropriate for human well-being. Conversely, the transgression of these boundaries could alter the current state of the Earth in an irreversible manner.

PBs have been established for climate change, change in biosphere integrity, stratospheric ozone depletion, ocean acidification, biogeochemical flows, land-system change, freshwater use, atmospheric aerosol loading, and the introduction of novel entities. Further details are discussed in section 3.2.1.

For all the boundaries proposed, currently, six out of nine planetary boundaries have been to be transgressed at the global level (i.e., climate change, biosphere integrity, land system change, biogeochemical flows of N and P, freshwater change, and novel entities). Boundaries and their current state can be found in Richardson et al. [35]

The boundaries for the different systems, as mentioned above, are based on data obtained from geosphere-biosphere interactions throughout the Earth's history. However, the introduction of novel anthropogenic entities has made it difficult to quantify their boundaries because many of their impacts and interactions with the Earth's systems remain unstudied. Because of this, mankind has often been surprised by the consequences of their unintended release and the effects on the integrity of the biosphere, as was the case with chlorofluorocarbons on the ozone layer. It is for this reason that not having the repercussions of the existing novel entities, the impacts of this boundary are usually not taken into account. Its application will be shown in more detail in section III.

1.4.3 Ex-ante LCA

Currently, LCA can be divided into two applications: Ex-post LCA studies and ex-ante LCA studies. The former focus on technological systems that are fully operational, studies. Conversely, ex-ante LCA focuses on evaluating emerging technologies before they are commercially implemented, i.e., when the technologies are still in the laboratory or at pilot scale, and therefore, system specifications and industrial scale data are not yet available. This is intended to improve technology development and/or to compare an emerging technology at scale with an existing technology as a reference. [36]

Ex-ante LCA does not predict the future. Rather, it explores the future by evaluating a range of possible scenarios that define the space in which the technology can operate. This will allow verification of design options that could steer the technology toward a preferred future state. Cucurachi [36] classifies the approaches employed in ex-ante LCA into five types (i.e., Prospective, Consequential, Dynamic, Anticipatory and Mixed), with the main difference lying in the specific focus of the analysis. Further information about ex-ante LCA can be found in section V.

Among these approaches, we can highlight the so-called prospective LCA (p-LCA), which aims to study future technological systems and their environmental implications. p-LCA needs to deal with the dynamic nature of technological advancements, which introduces complexities in anticipating long-term environmental impacts. Hence, one of the key challenges associated with p-LCA is the inherent uncertainty in predicting future scenarios accurately, for which p-LCA needs to make assumptions (more details will be provided in section.4.2.)

P-LCA, as evidenced by its advantages and disadvantages, emerges as a powerful tool for shaping sustainable practices and guiding decision-making in the medium and long term. While challenges exist, ongoing research and refinement of methodologies hold the promise of addressing these concerns, ultimately enhancing the effectiveness and applicability of p-LCA in fostering a more sustainable future.

Incorporating p-LCA into the decision-making process helps navigate uncertainties linked to emerging technologies. As sustainability becomes a central concern across various industries, the ability to anticipate and address environmental impacts becomes paramount. p-LCA aids in the identification of areas where improvements can be made, enabling the development and implementation of strategies that enhance overall sustainability (see section 4.3.). This forward-looking methodology contributes to a more resilient and adaptive approach, ensuring that future innovations align with ecological principles and contribute positively to global sustainability objectives.

1.5 Outline: problems addressed

The utilization of systematic modeling and optimization tools to address challenges within the transportation sector holds significant potential, offering

valuable insights to guide decision-makers and policymakers in formulating sustainable strategies. Below a brief summary of the three specific problems addressed in this thesis is provided, highlighting their integration of economic, environmental, and social aspects.

1.5.1 Navigating current options: Selecting the optimal biofuel alternatives (Article 1)

The continued growth in world energy demand is projected to increase by 12% between 2019 and 2030. Given that transport is a major player in energy consumption, still relying heavily on oil (i.e., covering 92% of its fuel demand), the current practices of the sector are deemed unsustainable.

This work explores the potential of biofuels, such as biodiesel and bioethanol, as promising alternatives to fossil fuels in the transport sector. With their ability to mitigate climate change and provide ancillary benefits to society, biofuels can contribute to a sustainable development.

While the advantages of biofuels are recognized, it is essential to acknowledge their negative side effects, especially competition for land and water use. This study delves into the production of biofuels, considering various feedstocks and processes, each of which contributes to different environmental impacts and engine performance. The objective is to identify the most suitable biofuels, and for those deemed inefficient, this work provides quantitative improvement targets that, if achieved, would make them efficient.

To address this, we propose the use of DEA, a non-parametric method for benchmarking alternatives that can combine multiple indicators into a single performance score and provide insights into improvement potential for inefficient alternatives compared to the best-performing processes.

The proposed approach evaluates the performance of 72 different biofuel routes, considering three sustainability dimensions (economic, environmental, and social) based on 12 different indicators. The analysis encompasses the entire life cycle of the biofuels, quantified via LCA, from cultivation and production to distribution and final combustion in vehicles (cradle-to-wheel). Figure I-4 shows a graphical abstract of article 1.

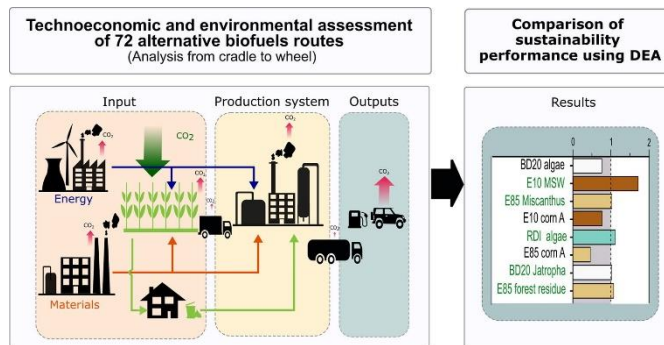


Figure I- 4. Graphical abstract of article 1: Comparing biofuels through the lens of sustainability: A data envelopment analysis approach

1.5.2 Designing an emerging process for optimal biofuel production for sustainable transportation.

Currently, global biofuel production relies on agricultural crop biomass (i.e., first generation biofuels), creating a competition for land between fuel and food production. To mitigate these challenges, the exploration of alternative sources, such as microalgae, holds promise. Microalgae has advantages like rapid growth and minimal land use, offering a sustainable alternative to conventionally farmed biofuels.

However, biofuel production from microalgae is an energy-intensive process, with significant impacts occurring upstream (e.g., energy production for water recirculation). Recognizing this, this thesis combines the principles of LCA and an AESA method based on PBs as I tools to distinguish the most sustainable technologies for microalgae-based biofuel production. Additionally, the study evaluates the potential of microalgae biofuels to diminish human health impacts compared to both, fossil fuels and conventional biofuels. Our study adopts a cradle-to-wheel perspective, focusing on three production process of biofuels (Transesterification, hydrodeoxygenation, and hydrothermal liquefaction), and considering different technological options for the carbon sources (i.e., direct air capture and natural gas power plants), the use of byproducts (i.e., residual biomass and biogas), and different electricity mixes (i.e., electricity mix in 2020 and 2040). Figure I-5 shows a graphical abstract of article 2.

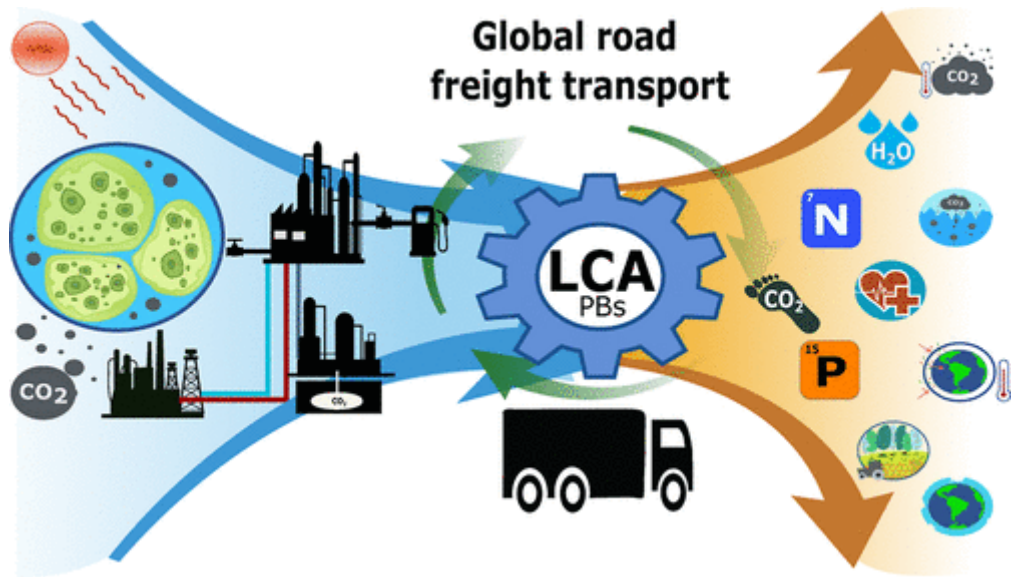


Figure I- 5. Graphical abstract article 2: Microalgae Biofuel for a Heavy-Duty Transport Sector within Planetary Boundaries

1.5.3 Prospective life cycle assessment for assessing transportation sector alternatives (Article 3)

Nowadays, the transportation sector is undergoing a critical transition, with many global policies aiming for gradual decarbonization through technological innovation. As an example, the European Commission, in alignment with the goal of achieving carbon neutrality by 2050, has set a target of a 40% reduction in emissions in the transportation sector by 2030.

To meet these ambitious climate objectives, the adoption of low-carbon fuels such as biofuels and electric vehicles (EVs) is deemed fundamental. However, many existing studies about their environmental performance assume a static scenario, neglecting the potential impact of improved efficiencies or the integration of new decarbonization technologies in the long term[37]. While these contributions offer valuable insights into the current environmental performance of transportation technologies, they may fall short of capturing the long-term implications of today's technological choices.

To address this gap, we employ p-LCA, which, opposite to traditional LCA, explicitly includes the effects of future changes in LCA datasets' background system (i.e., in all activities not explicitly modelled). This is done by generating

future life cycle inventories based on the results obtained from Integrated Assessment Models (IAMs) for various socioeconomic and environmental policies. Therefore, p-LCA considers potential changes that may influence environmental impacts over time, ensuring that long-term decisions are not made based only on today's assets.

In this section IV, we conduct a prospective life cycle assessment spanning the century, evaluating the global road freight transport (RTF) sector across 12 distinct regions. Our analysis incorporates diverse policy scenarios for rapid decarbonization, our primary focus centers on two alternatives: trucks powered by biofuels (bioICTs) and battery electric trucks (BETs)

In addition, we here depart from the conventional focus on a singular zone to study regional variations and their ultimate impact at the global level. Figure I-6 shows a graphical abstract of article 3.



Figure I- 6. Graphical abstract article 3: The future of sustainable road freight transport: when and where will electric trucks be more sustainable than biofuel powered trucks?

1.6 General conclusions

This thesis is devoted to contribute in addressing the evolving sustainability challenges within the transport sector and guiding future sustainability policies. To accomplish this, we have integrated LCA methodology within decision-making support tools. We next provide a set of conclusions that we accomplished in this thesis:

- The integration of DEA and LCA facilitates a comprehensive evaluation of various biofuel production alternatives, emphasizing sustainability considerations. This analysis leads to the conclusion that policies should prioritize the widespread adoption of renewable diesel over traditional ethanol or biodiesel, given its superior performance.

- To comprehensively assess the environmental impact of biofuel production, we employed an AESA method, utilizing the PBs metric. We consider microalgae biofuels as an emerging alternative for freight road transport, where some microalgae biofuel routes emerged as promising alternatives to fossil fuels, showing potential for climate change mitigation and reduced biosphere impacts compared to conventional biofuels
- Prospective Life Cycle Assessment (LCA) was employed to propose policies for the implementation of biofuels and electric trucks in road freight transport. This comprehensive analysis, considering the medium and long-term evolution of technologies, revealed the best alternatives for decarbonization in the studied regions worldwide until 2100. While in the long term, BETs are presented as a better alternative than bioICTS, BETs show higher GHG emissions compared to conventional diesel trucks in eight of the 12 regions analyzed through 2030, highlighting the continued importance of liquid fuels, especially biofuels.

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CHAPTER II

II. Navigating current options:
Selecting the optimal biofuel alternatives

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

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2.1. 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 [38–40].

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 in 2020 and 34.4 billion liters in 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 paraffin) 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 biofuel 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 using six possible fuels (i.e., ethanol, biodiesel, RD or HVO, diesel and gasoline) obtained from 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 biofuels, which will be deemed efficient. For non-suitable biofuels, 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.

2.2. 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.

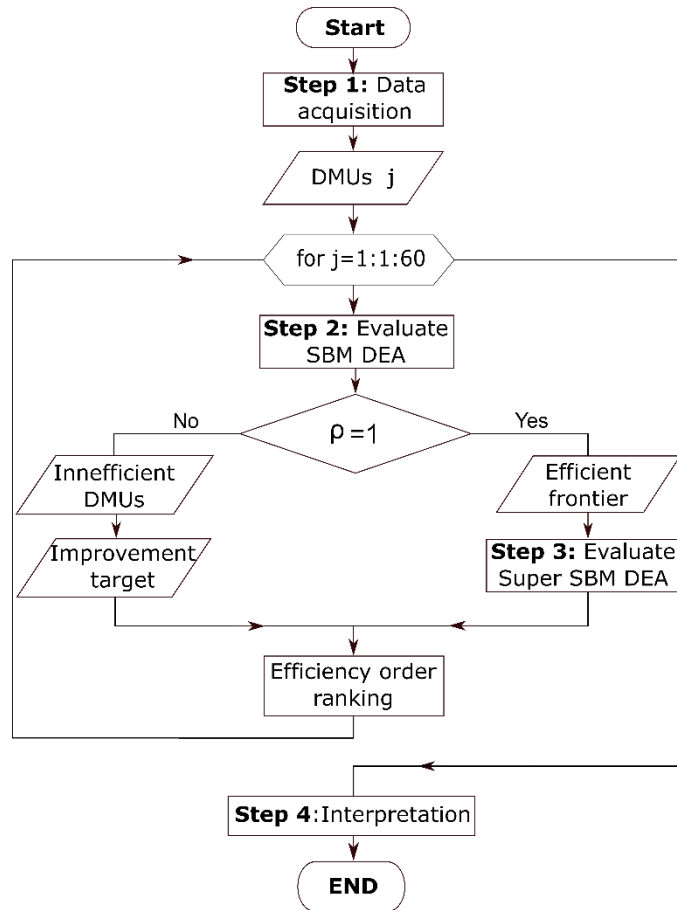


Fig. 1 Flowchart for the methodology proposed.

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.

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.

2.2.1. 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 (CID) 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 CID 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].

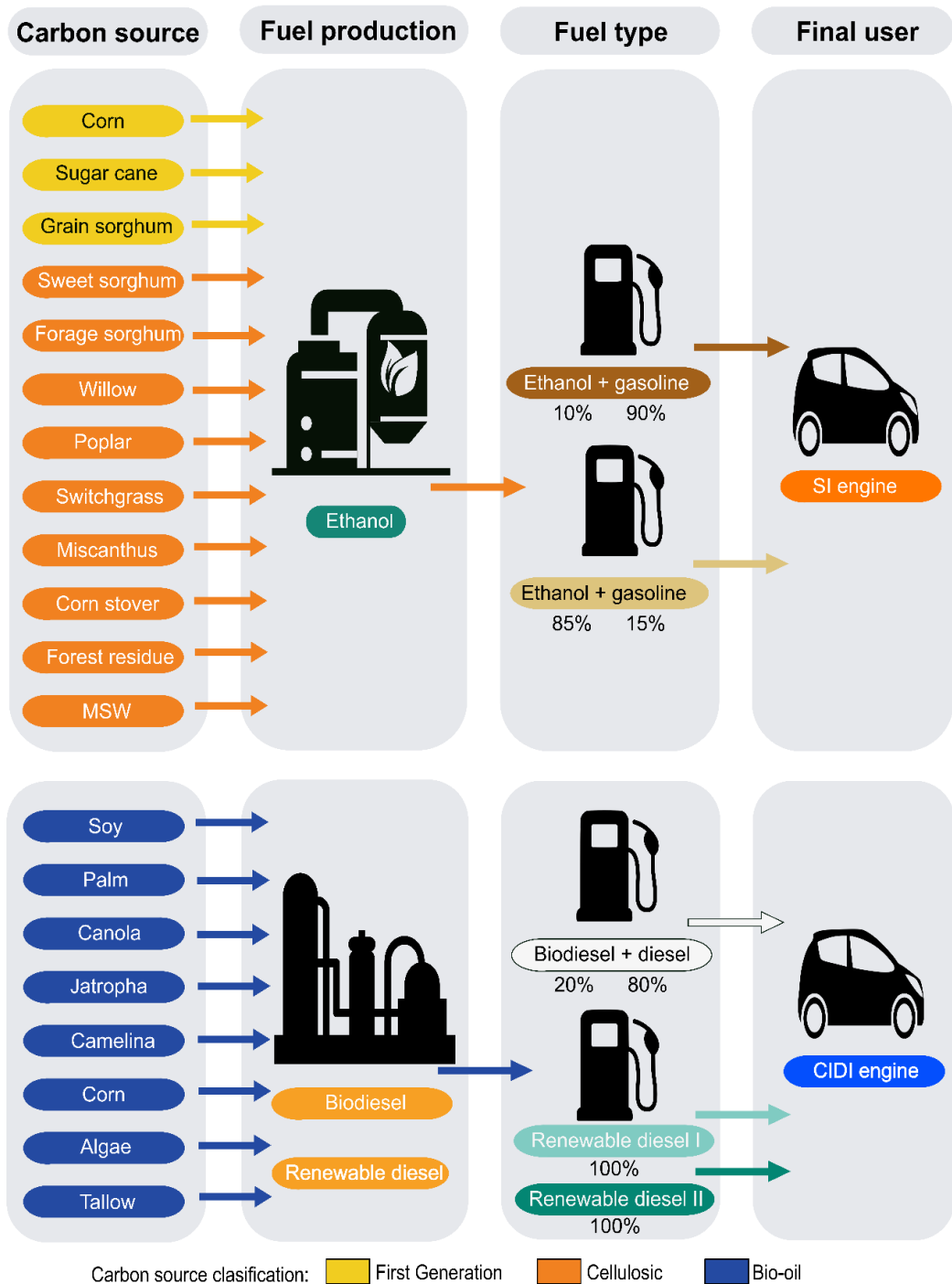


Fig. 2 Block-diagram providing the different alternatives considered as carbon source, fuel production process, blend and type of combustion engine. Carbon sources are depicted with a different color depending on whether they are first-

generation (e.g., corn), cellulosic (e.g., poplar) or bio-oils (e.g., palm). SI: Spark Ignition; CIDI: Compression Ignition Direct Injection.

Overall, 50 ethanol-based fuel routes are considered as follows. On the one hand, we study a total of 19 routes to produce ethanol through 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 standalone fuel, i.e., not blended with diesel).

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 Electronic 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 ($Input_{f,p}^{Raw}$):

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

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 Electronic 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.

$$Cost = \sum_{f,p \neq \text{farming}} Input_{f,p} UC_f \quad (2)$$

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 Electronic Supplementary Material.

$$Impact_u = \sum_p Impact_{u,p}^{Stage} \quad \forall u \quad (3)$$

$$Impact_{u,p}^{Stage} = \sum_f Input_{f,p} Ecovector_{u,f} \quad \forall u, p \quad (4)$$

$$= \{farm, transport, conversion\}$$

$$Impact_{u,p}^{Stage} = \sum_e Emission_{e,p} IF_{u,e} \quad \forall u, p = \{combustion\} \quad (5)$$

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 CO₂ absorbed during photosynthesis, thus resulting in a net-zero cycle. Therefore, the CO₂ 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 [41], while fuel densities are provided in Table S21 in the Electronic Supplementary Material.

Finally, social indicators (i.e., land occupation and water use) are obtained as follows. The land occupation indicator (*Land*, in [*ha·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 liter of biobased fuel (*Crop*, e.g., tons of poplar needed for 1 liter 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.

$$Land = \frac{Crop}{Yield^{Crops}} \quad (6)$$

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).

$$Water = Water^{Inputs} + Water^{Crops} \quad (7)$$

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

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

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 Electronic 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 Electronic Supplementary Material and summarized here in Table 1, where biofuels are grouped into five categories according to their production process.

Table 1 Statistics of the sustainability indicators considered for the 72 biofuel routes. Values are for 1 liter 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	4.30	4.34	1.82	2.10

	(4.28- 5.45)	(0.005- 5.23)	(0.004-7.91)	(1.28-6.15)	(1.50-7.29)
	2.58	0.67	4.58		
TE [10^{-3} kg 1,4-DCE]	(2.58- 2.63)	(0.005- 3.12)	(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; POFPP: photochemical oxidant formation potential; TA: Terrestrial acidification; TE: terrestrial ecotoxicity.

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 will be 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.

2.2.2. DEA Fundamentals

DEA [10] 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 ($i = 1, \dots, m$) into multiple outputs ($r = 1, \dots, k$) [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 $2 \cdot (m \cdot k)$, where $m \cdot k$ is the product of the number of inputs times the number of outputs [53][42]; such a condition is satisfied in the present analysis (i.e., $72 > 2(3 \cdot 9)$).

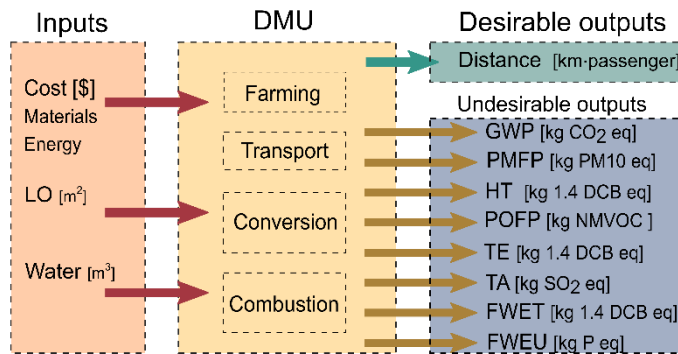


Fig. 3 Inputs and (desirable and undesirable) outputs considered for each biofuel (DMU). Units for each indicator are provided between brackets. LO: land occupation; Water: water used in farming plus water depletion; GWP: global warming potential; FWEU: freshwater eutrophication; FWET: freshwater ecotoxicity;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

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 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].

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-A₁-A₂, 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 A₁-B-A₂, provided that inputs are not allowed to worsen.

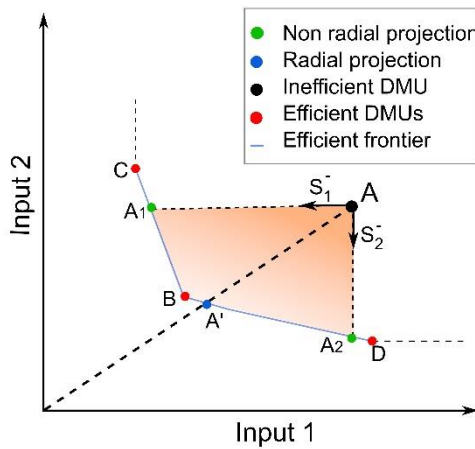


Fig. 4 Difference in projection onto efficient frontier by the radial and the non-radial SBM models

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 CO₂ emissions [72], the measurement of environmental efficiency of

transportation sector based on CO₂ emissions [73], and the relation between social fixed assets investment and GDP in the industry with SO₂ emissions [74]. The mathematical model is described in detail in the next section.

2.2.3. 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.

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

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

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

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

$$\lambda_j \geq 0 \quad j = 1, 2, \dots, n$$

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_{i0} and y_{r0} 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:

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

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

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

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

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

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: $\rho^* = \tau^*, \lambda^* = \frac{\Lambda^*}{t^*}, S^{-*} = \frac{s^{-*}}{t^*}, S^{+*} = \frac{s^{+*}}{t^*}$.

2.2.4. 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:

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

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

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

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

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 $\sum_{i=1}^n \lambda_i = 1$ and $\sum_{i=1, \neq 0}^n \lambda_i = 1$ in models (m.1) and (m.3), respectively.

2.2.5. 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).

2.3. 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] [43] and solved in an AMD Ryzen 5 4500U processor for each of the 72 DMUs in each scenario. Each instance took less than 1s 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 explicit mention to result distributions.

2.3.1. 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 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, owing 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 m³ of water/liter of E85 from MSW, compared to 0.76 m³ 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 liter of ethanol from first-generation biomass (i.e., fermentation of simple sugars) emits on average 0.47 kg CO₂ eq, while the production of 1 liter of ethanol from cellulosic materials and an acid hydrolysis process emits only an 0.21 kg CO₂ 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, 0.23kg of dry MSW is generated per day and person in Europe. If all this waste were used to produce E85, 0.08 liter 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 liter/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).

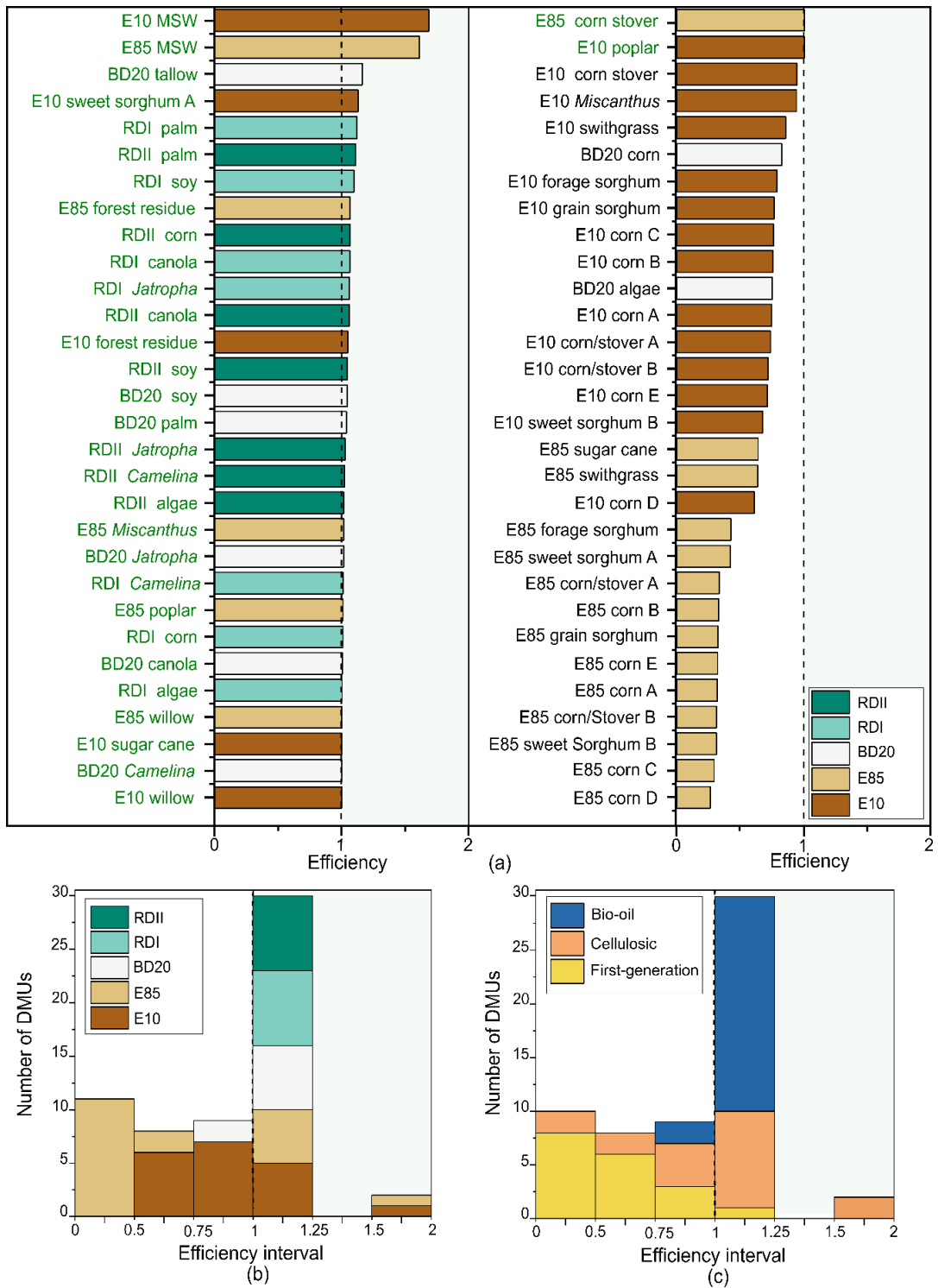


Fig. 5 Efficiency scores for biofuels. (Super)efficiency scores for the 72 biofuels routes are provided as horizontal bars in subplot (a), with biofuels sorted in decreasing order of efficiency and efficient biofuels depicted with a green label. Histograms at the bottom of

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

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 (Figure 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 (Figure 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 [44]. 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 km² [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., 194g of fertilizer per liter of corn-based ethanol, compared to 10g 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 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 (Figure 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.*, total GWP without deducting biogenic CO₂). 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 20% of biofuel blends will have a limited benefit on climate change, which calls for policies promoting pure biofuels or blends with higher biofuel content.

A totally different picture emerges when other environmental impacts are assessed. In the case of terrestrial ecotoxicity (Figure 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.

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.

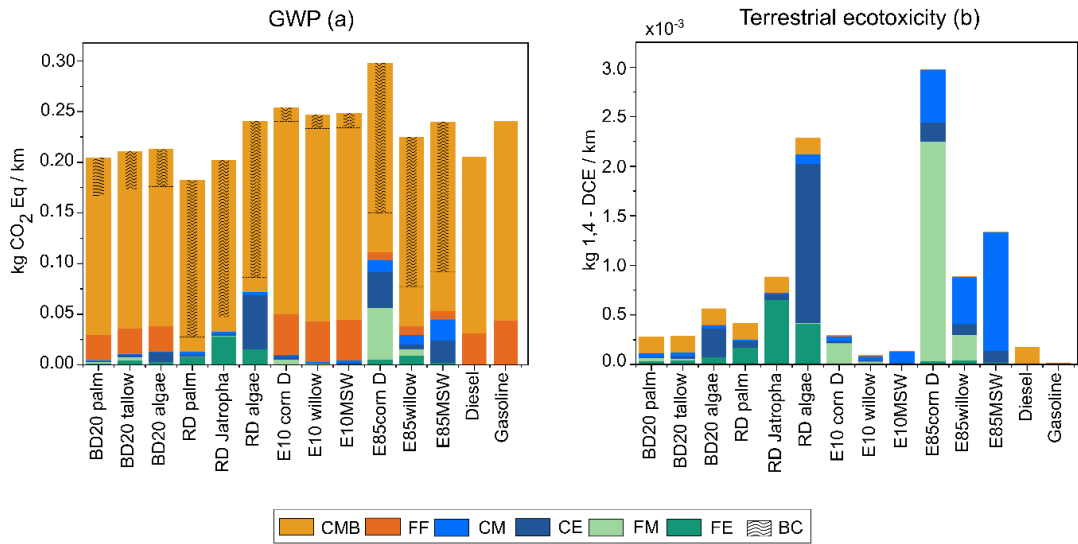


Fig. 6 Breakdown of the GWP (subplot (a)) and terrestrial ecotoxicity (subplot (b)) generated in the life cycle of selected biofuels. CMB: Combustion; FF: Fossil fuel production; CM: Materials for the biofuel production; CE: Energy for biofuel production; FM: Farming materials; FE: Farming energy; BC: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

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 [91]. 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).

2.3.2. 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 Electronic Supplementary Material, while detailed results for the nominal scenario are given in Table S31.

DMU	Inputs				Undesirable Outputs								Outputs
	Eff.	Cost	LO	Water	GWP	FWET	FWEU	HT	PMFP	POFP	TA	TE	Distance
BD20 algae	0.82	15%	35%	0%	8%	0%	61%	44%	18%	18%	0%	74%	0%
BD20 corn	0.75	0%	34%	72%	15%	0%	5%	7%	18%	18%	14%	9%	0%
E10 cellulosic	0.91	0%	33%	14%	2%	0%	2%	0%	0%	2%	2%	41%	0%
E10 cellulosic sorghum	0.73	6%	75%	83%	0%	9%	14%	12%	4%	7%	1%	79%	1%
E10 corn	0.72	0%	60%	96%	10%	12%	26%	19%	5%	6%	1%	70%	0%
E10 grain sorghum	0.32	0%	87%	27%	6%	18%	23%	20%	4%	5%	0%	70%	0%
E85 cellulosic	0.63	0%	0%	53%	29%	37%	50%	44%	31%	32%	50%	76%	0%
E85 cellulosic sorghum	0.39	42%	99%	99%	1%	4%	43%	45%	26%	46%	18%	97%	40%
E85 corn	0.31	1%	99%	98%	46%	40%	62%	63%	40%	47%	25%	98%	42%
E85 sugar cane	0.64	0%	0%	76%	21%	0%	52%	58%	43%	51%	43%	55%	0%
Average improvement		6%	52%	62%	14%	12%	34%	31%	19%	23%	15%	67%	8%
% decrease	0		20		40		60		80		90		100
% increase	0		20		40		60		80		90		100

Fig. 7 Improvement median targets for inefficient biofuels to become efficient considering 100 possible scenarios. E10 cellulosic: average improvement targets across E10 switchgrass, E10 Miscanthus & E10 corn stover; E10 cellulosic sorghum: average improvement targets across E10 sweet sorghum B & E10 forage sorghum; E10 corn: average improvement targets across E10 corn A-B-C-D-E & E10 corn/stover A-B; E85 cellulosic: average improvement targets across E85 switchgrass & E85 corn stover; E85 cellulosic sorghum: average improvement targets across E85 sweet sorghum A-B & E85 forage sorghum; E85 corn: average 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.

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 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 [92].

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 [93,94]. 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 [95]. Works by Castillo et al. [96] and Zhang et al. [97] [97] 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 [98,99].

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 (%) 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[100], 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 NO_x Trap, or Exhaust Gas Recirculation that reduces the combustion temperature [101]. 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.

2.3.3. 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 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.

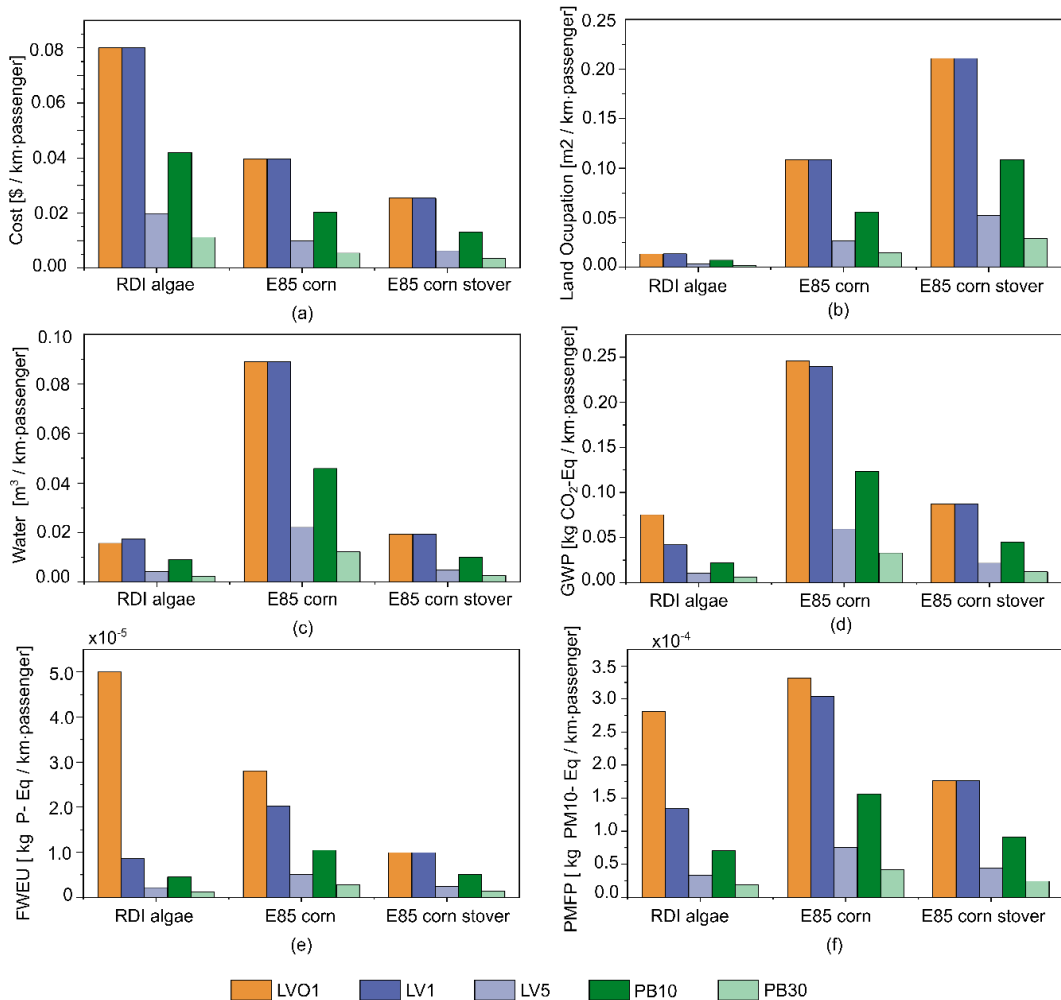


Fig. 8 Changes achieved in inputs (i.e., costs, land occupation and water use) and selected undesirable outputs (i.e., global warming potential, freshwater eutrophication and fine particulate matter formation) by replacing the current mix (i.e., US-WECC) and vehicle for three fuels with a different type of carbon source: 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).

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 [102] 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 [103] or the fermentation process [104] 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 [105,106], 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.

2.4. 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 prove 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.

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CHAPTER III

III. Designing an emerging process for optimal biofuel production for sustainable transportation

Microalgae Biofuel for a Heavy-Duty Transport Sector within Planetary Boundaries

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KEYWORDS: Microalgae, biofuels, LCA, planetary boundaries, renewables, biosphere integrity, human health.

3.1. Introduction

In a context where the transport sector is responsible for 22% of the global carbon emissions generated, 1 electric vehicle emerge as a promising alternative for sustainable mobility. While this alternative is suitable for low-range vehicles used for urban mobility, 2 the current state-of-the-art of batteries 3 limits fuel substitution in heavy-duty vehicles that tend to travel longer distances, making liquid fuels from bio-based feedstock promising candidates for reducing the environmental impact exerted by this sector 4.

Currently, the world production of biofuels is based on agricultural crop biomass causing competition for the available land between fuel and food production. 5,6 This exacerbates the risk of losing biodiversity and ecosystem services. Some of these problems could be avoided by producing liquid fuels from microalgae, which, compared to conventionally farmed biofuels, shows advantages such as rapid growth and low or marginal use of land. Carbon sequestration and the capacity of self-producing energy using byproducts are additional advantages. 7,8

Microalgae are carbon-fixing microorganisms that require adequate CO₂ concentrations to thrive, typically with CO₂ enriched air flow with concentrations in the range of 0.8 -10% volume. 9 Considering that the atmosphere contains a low concentration of CO₂ (i.e., around 0.041%), supply of atmospheric air would not be enough, and additional CO₂ should be injected to prevent an insufficient concentration that would limit productivity. CO₂ can be obtained by capture techniques, either used at point sources such as steam

methane reforming, NH₃ production, or natural gas power plants (NGP),¹⁰ or by using direct carbon capture from the air (DAC).⁸ In both cases, engaging microalgae production with CO₂ capture has the potential to reduce the negative effect on climate change compared to the use of conventional fuels, either by avoiding some carbon emissions at point sources or by resorting to atmospheric (instead of fossil) CO₂ when using DAC. Another interesting possibility is the recovery and storage of the carbon embedded into the microalgae byproduct, which otherwise would be released back into the atmosphere upon natural decomposition. This can be done by capturing the CO₂ emitted during the use of the residual biomass for cogeneration, and storing it in a geological deposit,¹⁰ thus converting the biofuel production process into a carbon capture utilization and storage process.

Among the different biomass conversion routes, the two routes that stand out are (1) the production of biodiesel (BD) from a solvent-based lipid extraction with a subsequent transesterification, and (2) the production of green (renewable) diesel.¹¹ In turn, green diesel can be produced by two processes: hydrodeoxygenation after lipid extraction (HDO) or hydrothermal liquefaction (HTL). For the HTL process, the production of biofuel does not require a previous lipid extraction process, allowing the use of feedstocks with up to 20% moisture content avoiding the use of dehydration and drying pretreatments

Biofuel production from microalgae is an energy-intensive process, where most impacts occur either upstream (e.g., energy production for water recirculation) or downstream (e.g., biofuel combustion in the vehicle engine) of the main facility. This calls for the application of life cycle assessment (LCA)¹² as an essential tool to identify the most sustainable technologies for microalgae-based biofuel production.

Since its development, LCA has allowed for comprehensive environmental analyses of processes and products, covering activities from feedstock extraction to waste management. So far, several impact assessment methods have been put forward (e.g., CML, ReCiPe), yet they lack absolute thresholds to elucidate whether a given system should be deemed sustainable. To overcome this limitation, here we focus on an absolute environmental sustainability assessment (AESAs) method, which bridges conventional LCA principles with the concept of Planetary Boundaries (PBs), developed by Rockström et al.¹³ and Steffen et al.¹⁴ The PBs framework aims to quantify the absolute environmental sustainability level of human activities by proposing nine bio-geophysical boundaries for the Earth system that define a "safe operating space for humanity" (SOS). These boundaries represent quantitative thresholds whose transgression could alter the current state of the Earth in an irreversible manner.¹⁴ So far, planetary boundaries have been established for climate change, change in biosphere integrity, stratospheric ozone depletion, ocean acidification, biogeochemical flows, land-system change, freshwater use, atmospheric aerosol loading, and the introduction of novel entities. The transgression of

the SOS undermines the resilience of ecosystems that support human well-being and has repercussions on human health problems at different scales. 14

In the context of biofuels, several studies applied LCA to evaluate different production technologies. Most of them use a cradle-to-tank scope, whereby the focus is on the production of biofuels through transesterification processes, including also upstream activities (e.g., biomass production) but totally excluding the combustion stage. [12,13] Some of these works focus only on particular processes, such as microalgae production, 15 microalgae oil production,16 or fuel production 17, where the carbon source, and its downstream repercussions are mostly ignored. The LCA scope was extended in other contributions by Batan et al. 18 and Longwhen et al., 19 applying a cradle-to-wheel approach. However, these works focused only on greenhouse gas (GHG) emissions, thereby neglecting other relevant impacts such as land or water use. Given that electricity consumption and CO₂ supply are among the most operationally and economically significant factors in microalgae biofuel production, we include them in the present work. Yue et al. 20 and Bennion et al. 17 already explored these factors, but unlike them, we look into the use of renewables for electricity generation, which could bring additional environmental advantages.

For the first time, this contribution studies the transformation of microalgae into fuels through the lens of the planetary boundaries. Notably, we consider three different types of fuels (BD, HDO and HTL) under 68 scenarios combining different carbon sources (i.e., CO₂ from a natural gas power plants (NGP) and DAC), byproduct treatments (i.e., biomass combustion, and biogas production and combustion), and electricity mixes (i.e., 2020 global mix, electricity mix projected under the sustainable development for 2040), reaching a level of breadth in the analysis way above previous works We combine the principles of LCA and an AESA based on the PBs, adopting a cradle-to-wheel perspective. Our study goes beyond GHG emissions, embracing other impacts on key Earth-system processes, while also covering impacts on human health. Overall, this work evaluates the potential of microalgae biofuels to reduce environmental and human health impacts compared to both fossil fuels and conventional biofuels, with particular attention to impacts on Earth-system processes that are currently transgressed by anthropogenic activities (i.e., climate change, biosphere integrity, land-system change, and biogeochemical flows).

3.2. Methodology

The present study focusses on the environmental sustainability assessment of the production of biofuels from microalgae and their use in the heavy-duty transport, always considering a cradle-to-wheel perspective (i.e., thus including the combustion phase.

The 68 transformation scenarios of microalgae to biofuels are based on different combinations of alternatives for four technological decisions related to: (i) the source of CO₂, (ii) the use of byproducts such as Lipid-extracted algae (LEA), CO₂, gasoline, electricity or thermal energy, (iii) the electricity mix, and (iv) the type of fuel produced, as shown in Figure 1.

The first technological decision affects the selection of a source for the CO₂ that will be supplied to the microalgae to satisfy the carbon requirements and ensure that microalgae will never be CO₂-limited throughout the course of cultivation requirement. Although the literature suggests that lower CO₂ concentrations result in higher lipid production, 21,22 we consider compression and pumping of pure CO₂ for consistency with the approach followed in the GREET database. This can come either from DAC or from NGP.

The second technological decision concerns the potential utilization of the byproduct generated during the extraction phase of BD and HDO process, the so-called , lipid-extracted algae (LEA). Three main alternatives are considered for LEA: (i) combustion to partially supply heat and electricity for self-consumption; (ii) production of methane using an anaerobic digester with subsequent biogas combustion; and (iii) disregarding the utilization of LEA. For this last option, we assume that the CO₂ embedded in the LEA as biogenic carbon would be released back into the atmosphere as a result of natural decomposition. Conversely, the first two options generate CO₂ as a byproduct during LEA or biogas combustion, which can be exploited in three different ways: (i) capture and utilization (CCU) in the cultivation process as a source of CO₂, (ii) capture and storage in geological reservoirs (CCS), and (iii) direct emission to the atmosphere. Overall, this translates into seven practical alternative pathways for byproducts in BD and HDO routes. In the case of HTL scenario data extracted considers a wood-microalgae mixture 23, CO₂ is the only byproduct generated, for which we consider the same three pathways as for the CO₂ stemming from LEA or biogas combustion, i.e., CCU, CCS, or direct emission (i.e., no capture).

The third technological decision is related to the electricity mix that will supply the electricity requirements of the processes explicitly modeled in the study (i.e., foreground processes only). In this regard, two scenarios are considered: the 2020 global electricity mix (M2020) and sustainable global mix for 2040 (M2040). 24 This technological decision is of utmost importance, not only because the production of algae oil fuel is intensive in the electricity consumption (6.37 MJ/l of HDO; compared to 0.29MJ/l of HDO for soybean-based fuel), but also because the decarbonization of the electricity mix might potentially decrease impacts on the control variables of the PBs. 25

Finally, the fourth technological decision is devoted to the transformation of microalgae into a particular type of biofuel. Options considered include: (i) biodiesel production from

a solvent-based lipid extraction, with a subsequent transesterification and posterior blending with diesel in a 20% v/v blend (BD20), (ii) renewable diesel production by hydrodeoxygenation after the lipid extraction, and (iii) renewable diesel based on hydrothermal liquefaction of the microalgae.

Different alternatives for cultivation, drying, and extraction technologies were also considered at an earlier stage of the study (see Table S31) but were disregarded due to their low Technology Readiness Levels (TRL) and are not included in the aforementioned scenarios. The alternatives selected for these processes correspond to the technologies with the highest TRL, namely, open pond technology for microalgae cultivation, centrifugation and flocculation for microalgae drying, and wet solvent extraction for the oil extraction process.

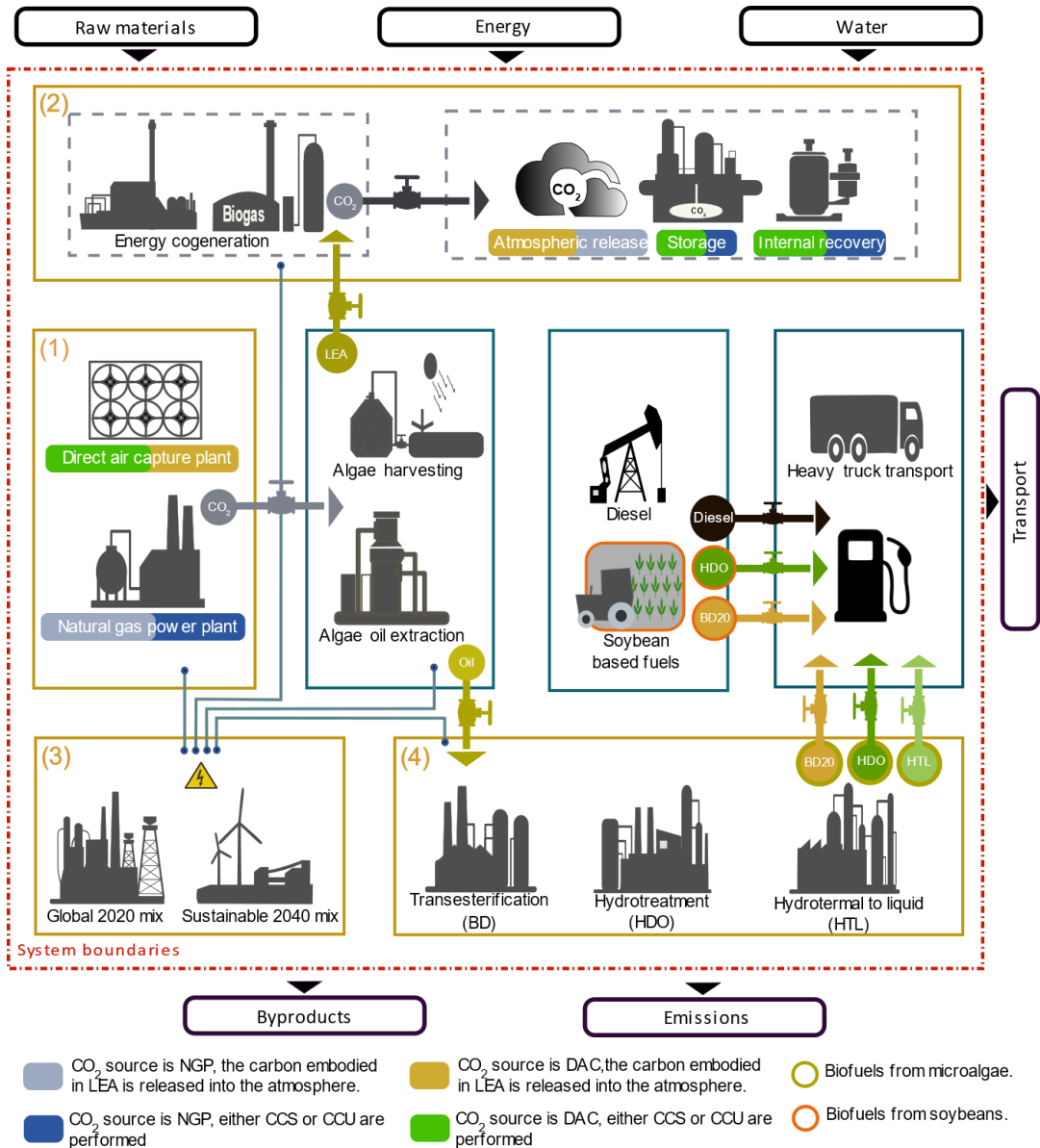


Figure 1. Conceptual framework for biofuels production from microalgae considering 68 scenarios based on four technological decisions: (1) Carbon feedstock from carbon captured from DAC and a power plant; (2) Carbon storage, carbon utilization, and carbon emissions considering the end life of CO₂ from lipid extracted algae (LEA); (3) 2020 global electricity mix, and sustainable 2040 electricity mix; (4) Transesterification, hydrotreatment and hydrothermal to liquid as the biofuel conversion process.

Figure 1 summarizes the 68 scenarios for microalgae-based biofuels combining different alternatives for CO₂ sources (two); the electricity mix (two); and potential routes for the exploitation of LEA and/or CO₂ byproducts (seven in the case of BD and HDO, and three for HTL). We also consider three additional scenarios for the sake of comparison,

consisting of diesel from fossil sources for heavy-duty transport, which we label as the business-as-usual (BAU) for diesel scenario, and biofuels based on soybean (i.e., BD and HDO). The addition of these three scenarios to the microalgae scenarios adds up to 71 scenarios in total.

For the sake of readability, the different scenarios will be named after colors according to the origin of the CO₂ supplied to the microalgae and the management of the CO₂ byproduct as follows. GREY will be used when the CO₂ source is NGP and the carbon embodied in LEA is released into the atmosphere, BLUE when the CO₂ comes from NGP and either CCS or CCU are performed, YELLOW when CO₂ comes from DAC and the carbon embodied in LEA is released into the atmosphere, and finally GREEN when CO₂ comes from DAC and CCS or CCU are performed. These colors will be complemented with a superscript related to the electricity mix (i.e., M2020 or M2040), and a subscript associated with the management of the CO₂ byproduct as follows: ACR when CO₂ is released to the atmosphere after cogeneration of LEA, CCU when CO₂ is reused in the cultivation process, CCS when CO₂ is stored in a geological reservoir, if CCU or CCS is not considered when there is no LEA cogeneration, CO₂ contained in the LEA is released into the atmosphere considering its natural decomposition. Finally, the color label will be accompanied by a suffix describing the type of biofuel production process (i.e., HDO, BD, or HTL). Although two cogeneration scenarios are considered (combustion and biogas production), to facilitate the discussion, only scenarios considering cogeneration by combustion are described in the main manuscript, leaving scenarios from biogas combustion in the Supporting information.

We acknowledge that such a large number of scenarios could motivate the use of optimization-based approaches to identify the ones showing a Pareto optimal performance.⁵ However, we preferred to retain the exhaustive analysis of all the scenarios to elucidate also those achieving a similar performance to optimal cases. These scenarios, although suboptimal under the assumptions considered, could become optimal under different conditions for plant location, season, or distances of CO₂ sources and storage sites relative to the biofuel production facility.

3.2.1. LCA and planetary boundaries

LCA quantifies the environmental impacts of products, processes, and services over their entire life cycle, covering a wide range of potential damages. To apply it, we follow ISO 14040 and 14044 standards based on four steps for identifying environmental hotspots. 12,26

The first LCA phase defines the goal and scope of the study. The goal of this environmental assessment is to quantify the absolute environmental sustainability level of the different scenarios for microalgae-based biofuel routes. To this end, we defined the annual world

ton-km (tkm yr⁻¹) demand for road freight activities as the functional unit, considered equal to 35 trillion tkm yr⁻¹ for 2022. 27 We adopt a cradle-to-wheel scope, thus covering all activities upstream of fuel production, the production of fuels and byproducts, and the final use of the fuel in vehicles intended for road freight activities (i.e., long haul-heavy trucks, 17t). An economic allocation was used as the attributional method to allocate impacts among the different products generated by each activity.

The second phase of the LCA quantifies the main inputs and outputs (i.e., energy, raw material, byproducts, and emissions) crossing the system boundaries. Here, mass and energy balance information from previous studies were retrieved for activities in the foreground system, namely, carbon sequestration, microalgae cultivation, microalgae drying, byproduct recovery, fuel production, and fuel combustion. Data for cultivation and drying phases were obtained from previous studies,^{23,28} considering freshwater use for farming activities as culture media. Material and energy requirements for the oil extraction phase were obtained from the GREET database,²⁸ Data source corresponds to harmonized values obtained from a microalgae production model that considers three types of algal strains: *Chorella Sorokiniana*, *Kirchneriella cornuta* and *Scenedemus obliquus*.²⁹ Similarly, mass and energy requirements for CCS processes were modeled according to previous works. 30–33 Then, this information was combined with the corresponding background activities data in ecoinvent v3.7.1 ³⁴, using SimaPro v11, ³⁵ to calculate the life cycle inventories (LCIs) of the different scenarios modeled. Additional details on the modeling of these scenarios are provided in section 2 of the Supporting information.

The third phase of the LCA involves assessing the damage produced by the LCIs in different environmental categories. To this end, nine control variables referring to seven Earth-system processes were considered.¹³ Aerosol loading and novel entities were omitted as these Earth-system processes PBs are not yet quantified.¹⁴ Hence, considering a set B of nine control variables of the PBs and a set S of 71 scenarios, the environmental impact caused by each scenario $s \in S$ in each control variable $b \in B$ ($IMP_{b,s}$) was calculated according to Equation 1.

$$IMP_{b,s} = \sum_{e \in E} LCI_{e,s} \cdot CF_{b,e} \cdot PV \quad \forall b \in B, s \in S \quad (1)$$

Here, $LCI_{e,s}$ represents the elementary flow e linked to the transportation of 1 ton of load across 1 km of distance through the use of the heavy-duty truck in scenario s . Elementary flows are referred to exchanges between the biosphere and the technosphere (e.g., kilograms of CO₂ fossil emitted). The characterization factor $CF_{b,e}$ computing the impact caused on control variable b by elementary flow e were taken from Ryberg et al. (36) for

all control variables of PBs except for biosphere integrity. For biosphere integrity, we used the characterization factors developed by Galán-Martin et al. 37, considering two main stressors of biodiversity loss, i.e., direct land use and CO2 emissions. Finally, the product between LCI and CF is, in turn, multiplied by the global road freight activities estimated for 2022 (PV, in tkm yr⁻¹) to determine the total impact linked to the functional unit.

Note that while PBs are defined at the planet level, variable $EB_{b,s}$ considers only the impacts from a particular economic sector (i.e., heavy-duty transport sector). To harmonize this difference in scope, different downscaling approaches (e.g., egalitarian, utilitarian, acquired rights or prioritarian) 38 can be used to assign a share of the whole SOS to the specific system under study.³⁹ However, sharing principles remain controversial, and there is no universal agreement on which or how they should be applied in practice.

Here, instead of using downscaling methods, we follow previous works and simulate the global anthropogenic impact of the whole economy (IMP^{GLO}) that would result from replacing the BAU scenario of the heavy-duty transport sector ($IMPT^{BAU}$) by an alternative biofuel scenario $IMPT^{ALT}$ 37,40. This is done by departing from the total current anthropogenic impact of the whole economy (EB^{CUR}), subtracting from it the contribution of the sector under study in the BAU scenario, and then adding the impact of the alternative scenario for the same sector (Equation 2).

$$IMP_{b,s}^{GLO} = IMP_b^{CUR} - IMPT_b^{BAU} + IMPT_{b,s}^{ALT} \quad \forall b \in B, s \in S \quad (2)$$

Here, $IMP_{b,s}^{GLO}$ is the global impact of the whole economy in control variable b under scenario s ; IMP_b^{CUR} corresponds to the current anthropogenic impact level in control variable b (after subtracting the natural background level, as shown in Table S2); IMP_b^{BAU} is the impact of the BAU scenario in control variable b ; and $IMPT_{b,s}^{ALT}$ is the impact of scenario s in control variable b .

We stress that in absolute sustainability studies, results are normalized relative to the maximum allowable impact to explicitly address the question of whether a system is environmentally sustainable in absolute terms in a given Earth-system process. Hence, with the global impact achieved in each scenario s at hand, we then calculate the PB footprint (PBF_s) by comparing the global (predicted) anthropogenic environmental impact ($IMP_{b,s}^{GLO}$) with the SOS_b as defined by Steffen et al. 14, using Equations 3 and 4.

$$PBF_s = \frac{\sum_b LT_{b,s}^{GLO}}{|B|} \quad \forall s \in S$$

$$LT_{b,s}^{GLO} = \begin{cases} 0 & \text{if } \frac{IMP_{b,s}^{GLO}}{SOS_b} < 1 \\ \frac{IMP_{b,s}^{GLO}}{SOS_b} & \text{otherwise} \end{cases} \quad \forall b \in B, s \in S$$

Here, the global level of transgression ($LT_{b,s}^{GLO}$) shows the fraction of the SOS occupied by the whole economy for control variable b under scenarios s for the global transport sector. PBs not transgressed after sector substitution will show an $LT_{b,s}^{GLO}$ value of 0. To determine the level of transgression caused only by the transport sector ($LT_{b,s}^{TRA}$), $IMPT_{b,s}$ will be used instead of $IMP_{b,s}^{GLO}$ in eq. (4).

Finally, in step four of the LCA methodology, results are interpreted, and recommendations are made. In this case, we analyzed the LTs of the scenarios to identify the main hotspots, comparing their absolute environmental sustainability performance, and determining whether they are truly sustainable. Section 3 of this manuscript is dedicated to this LCA phase.

3.2.2. Human health impacts

Although it is recognized that human health depends on safeguarding the natural systems that support human well-being, 41 the PB framework omits impacts affecting human health directly. For this reason, as done in previous works, 42 we complement our study by including the “Human health” (HH) impact category from the ReCiPe 2016 method. 43 In particular, we use the hierarchic perspective, which integrates impacts over a 100-year time horizon thus quantifying the HH endpoint indicator in terms of Disability-Adjusted Life Years (DALYs).43,44 This endpoint indicator considers the following midpoint metrics: global warming, ozone formation, stratospheric ozone depletion, fine particulate matter, water consumption, ionizing radiation, human non-carcinogenic toxicity and human carcinogenic toxicity.

3.2.3. Carbon footprint and CO2 balance

In addition to the PBs, we include in the analysis the well-known Carbon Footprint (CFP), which measures the total GHG emitted by a product over its life cycle, expressed in Gt of CO2 eq. Specifically, we follow the ReCiPe 2016 methodology for a time horizon of 100 years with the aim of comparing the results obtained from ReCiPe methodology with the results from the corresponding control variable for the PBs framework (i.e., atmospheric CO2 concentration and Earth's energy imbalance).

In order to calculate the CO2-based PBs control variables and the CFP, the benefits of CO2 uptake during biomass growth need to be properly assessed. This is a controversial issue,

45 since this uptake could be deemed as negative emissions depending on the system boundaries.

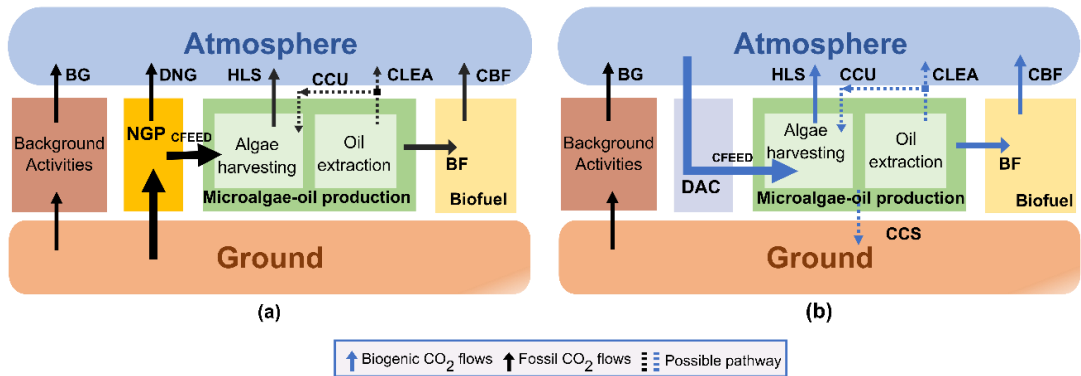


Figure 2. Carbon dioxide balance approach: (a) CO₂ captured from fossil point sources, b) CO₂ captured from direct air capture. NGP: CO₂ flow from the natural gas power plant; DAC: CO₂ flow from the direct air capture system; CCU: CO₂ from carbon capture and utilization from NGP; CLEA: CO₂ flow linked to the emissions from the combustion of lipid extracted algae residue; DNG: direct CO₂ emissions from NGP; CFEED: CO₂ flows for microalgae growth; HLS: CO₂ losses from algae cultivation; BK: CO₂ flow as emissions from background activities supplying materials and energy; CCS: CO₂ flow for storage; BF: CO₂ embodied into biofuel; CBF: CO₂ flow as emissions from biofuel combustion.

To perform a CO₂ balance in the atmosphere, we first classify CO₂ flows depending on the carbon source: (a) CO₂ captured from fossil point sources (NGP), and (b) CO₂ captured from direct air (DAC), as shown in Fig 2. These CO₂ flows are initially used for microalgae growth (CFEED), where part of this carbon flow will be released into the atmosphere owing to leakages in open ponds (HLS), while the rest will be incorporated into algae biomass. In turn, part of this carbon will remain embodied in the final biofuel until it is burned (CBF), while the rest will be part of the so-called LEA byproduct.

In our system, LEA can be burnt for electricity production and thermal energy for self-consumption. During this combustion, its carbon content will be released again as CO₂, which can be partly captured and stored in a geological reservoir (CCS). For direct combustion of LEA, around 82% of the CO₂ embodied is recovered, while in the case of biogas combustion, only a 33% can be retained. The remaining CO₂, which is emitted to the atmosphere, is labelled as CLEA in the figure. Alternatively to CCS scenario, we studied the option of reintroducing the CO₂ captured during LEA combustion into the cultivation process (CCU), thus decreasing the initial required flow of CFEED.

Finally, the CO₂ emissions which come from the background activities related to the system we explicitly modelled, are labelled as BG.

Overall, the expressions used to perform the CO₂ balance over the atmosphere are:

$$\sum_{e \in E} LCI_{CO_2, s} = \sum_{bg \in BG} LCI_{CO_2, bg, s} + \sum_{fr \in FR} LCI_{CO_2, fr, s} \quad \forall s \in S \quad (5)$$

$$\begin{aligned} \sum_{fr \in FR} LCI_{CO_2, fr, s} & \quad (6) \\ & = LCI_{CO_2, s}^{DNG} + LCI_{CO_2, s}^{HLS} + LCI_{CO_2, s}^{CLEA} + LCI_{CO_2, s}^{CBF} + LCI_{CO_2, s}^{CCS} \quad \forall s \\ & \in S \end{aligned}$$

where $LCI_{CO_2, bg, s}$ denotes the CO₂ flows from the background activities, $LCI_{CO_2, fr, s}$ represent the CO₂ flows from the foreground activities related to biofuel production, $LCI_{CO_2, s}^{BF}$ is the CO₂ embodied in the biofuel (stemming from the microalgae), $LCI_{CO_2, s}^{HLS}$ are the direct CO₂ emissions incurred during farming, and $LCI_{CO_2, s}^{CLEA}$ is the carbon embodied in the LEA, that can either be stored as CO₂ in geological reservoirs after LEA combustion, or HTL process ($LCI_{CO_2, s}^{CCS}$). In the case of CCS, since CO₂ is removed from the atmosphere, this corresponds to a negative flux whose sign is considered when making the calculations. The direct CO₂ emissions from NGP ($LCI_{CO_2, s}^{DNG}$) presented in Figure 2 are properly allocated between electricity and CO₂ based on economic allocation (see Table S15 in the Supporting information)

In the case of using CO₂ from DAC (Figure 2b), since the carbon used to grow the microalgae already comes from the atmosphere and no additional fossil carbon is required, $LCI_{CO_2, s}^{DNG}$ will not be accounted for. Note that the net CO₂ in the atmosphere ($\sum_{e \in E} LCI_{CO_2, s}$) will remain positive in both cases, since contributions from the background processes ($\sum_{bg \in BG} LCI_{CO_2, bg, s}$) will offset negative emissions in the foreground ($\sum_{fr \in FR} LCI_{CO_2, fr, s}$).

3.3. Results and discussion

The following section summarizes the results obtained for the different scenarios, that is, the impacts incurred by the different pathways of microalgae-based biofuels on different environmental categories (the PBs, the PBF, the CFP and HH). For benchmarking purposes, we also show the results of the BAU and two conventional biofuel alternatives for heavy-duty transport, namely, conventional diesel and biofuels based on soybean oil.

Relative contribution to the safe operating space

Figure 3 summarizes the level of transgression that would result from replacing the current transport sector with each of the different alternatives, expressed as the percentage of the SOS occupied by the heavy-duty transport sector in the nine PB control variables addressed. This level of transgression (i.e., variable $LT_{b,s}^{TRA}$) is calculated considering the lower, i.e., more stringent, limit of the uncertainty zone proposed by Steffen for the PB of every control variable. [24] In turn, the color indicates the level of transgression of the control variable, green when $LT_{b,s}^{TRA}$ is below 100%, yellow when $LT_{b,s}^{TRA}$ is larger than one but within the uncertainty region for control variable b , and red when $LT_{b,s}^{TRA}$ exceeds the relaxed PB proposed for control variable b (see Table S3 for control variable current values, along with their proposed PB). In addition, the PBF, CFP, and HH impacts are also provided for each scenario. Note that, due to space limitations, we only provide here the results for the most representative 12 microalgae biofuel scenarios. The remaining 56 scenarios for HDO, BD20 and HTL are available in section S3.1 of the Supporting information.

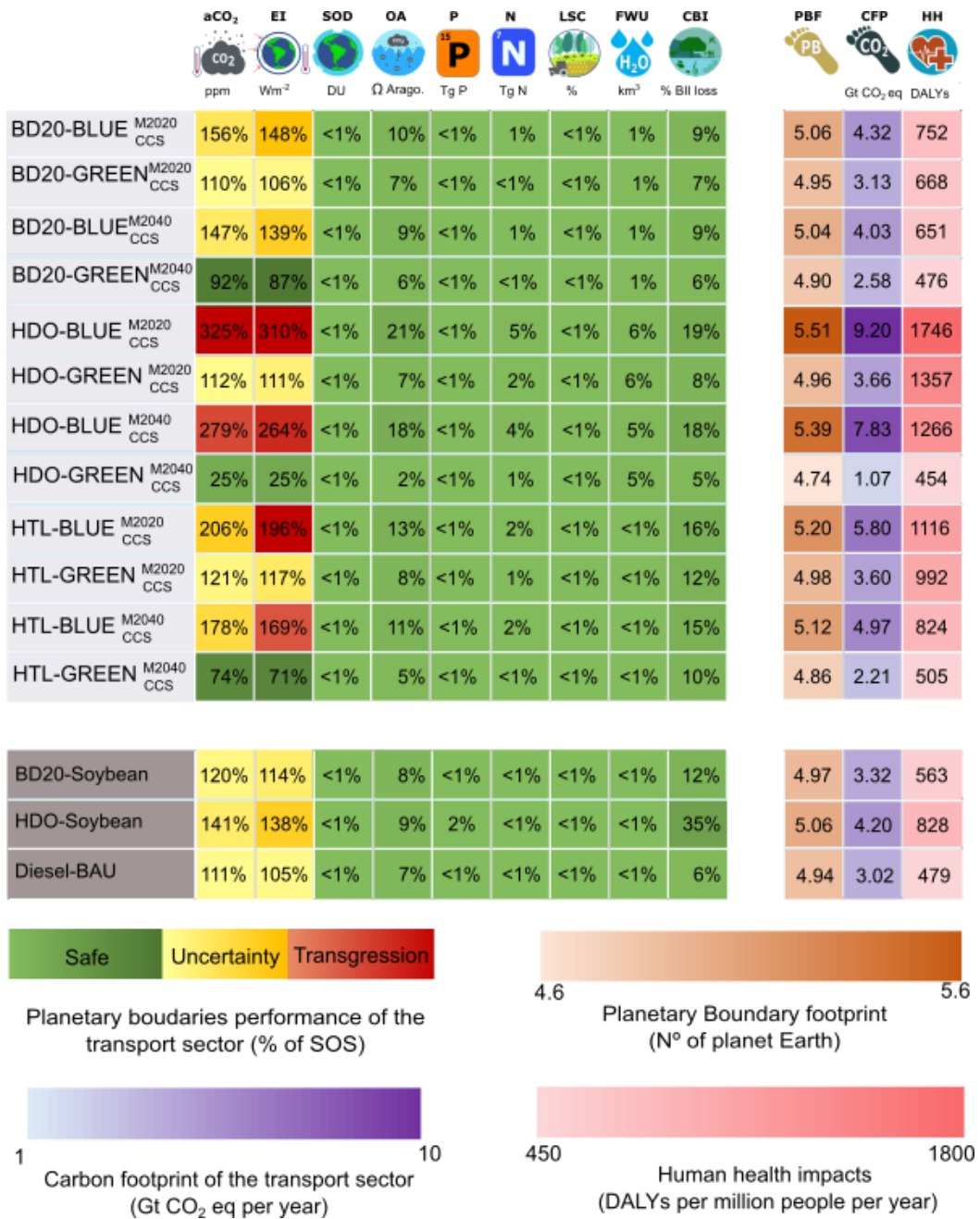


Figure 3. Impacts incurred by different scenarios for the global heavy-duty transport sector. Each row in the heatmap corresponds to a different scenario, as described by the labels in the first column: the fuel type (technological decision 4 in Fig. 1) and the alternative selected for technological decisions 1, 2 and 3, respectively (e.g., CO₂ source, electricity mix, and byproduct pathway). The remaining columns correspond to different impact categories as follows. The columns in the leftmost heatmap provide the level of transgression of the global SOS occupied by the heavy-duty transport sector alone. Then, the three columns, which are slightly separated, provide (from left to right) the PBF, the CFP, and the HH. Each set of metrics uses a different color key according to the four bar legends, where the corresponding units are also indicated. Acronyms for

the scenario labels are as follows. HDO: Renewable diesel 100% Vol from HDO; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; CCS: Carbon capture and storage in a geological reservoir; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed (aCO₂: atmospheric CO₂ concentration; EI: energy imbalance at the top of the atmosphere; O3D: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow-global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; PBF: Planetary boundary footprint; CFP: Carbon footprint; HH: Human health).

3.3.1. Conventional fuels and their impact on heavy-duty transport sector

According to the results obtained, the present heavy-duty transport sector (i.e., fueled by diesel) transgresses by a factor of 1.11 the SOS for CO₂ concentration. Results are not better for conventional soybean-based biofuels, which transgress the same boundary by a factor of 1.20 or 1.41, depending on whether soybean is used to produce BD or HDO. This suggests that the higher the percentage of biofuel in the final blend (20%v/v for BD20 vs 100% v/v for HDO), the greater the impact associated with CO₂ emissions. Note that we drop the label 100% to describe HDO and HTL scenarios, since none of these scenarios uses a different blend.

These results show that the current heavy-duty transport sector alone transgressed the SOS of climate change for the whole economy and that the scenarios where soybean is used as biofuel do not decrease the transgression but rather increase it even further. This is illustrated in the PBF metric, where the global transgression achieves values of 4.97 and 5.06 for BD20 and HDO, respectively; even higher than that for the BAU (diesel) scenario (4.94). This is due to the high impact on climate change during the production of soybean oil, and the low energy return on investment ratio obtained compared to fossil fuels (i.e., 20:1 for diesel compared to 2:1 in biodiesel).⁴⁶

Although the use of fuels based on soybean in the heavy-duty transport sector does not transgress the SOS for the remaining PBs (e.g., stratospheric ozone depletion or ocean acidification), their contribution to the change in biosphere integrity is relevant (i.e., 12% of the global SOS for BD20 and 35% for HDO), leaving little room for the remaining economic sectors. These results highlight the importance of finding alternatives that can reduce the impact on several PBs concurrently. With this spirit, scenarios for biofuels from microalgae will be discussed next.

3.3.2. Technological decisions on the production of biofuels from microalgae.

Considering the implications of the first technological decision, i.e., the carbon capture technology used for the CO₂ feedstock, the advantage of using DAC stands out (i.e., comparing GREEN vs BLUE). Despite capturing a certain amount of CO₂ with DAC requires

1.6 times more energy than doing it from a natural gas power plant owing to the lower concentration of CO₂ in the air compared to point sources, 47 DAC emits up to 2.9 times less fossil CO₂ in the life cycle. This is because, under the assumptions adopted in this work, all the CO₂ from NGP is modelled as fossil carbon and, therefore, it is a positive flow that increases the atmospheric CO₂ concentration. Hence, DAC-based pathways show a substantial improvement in terms of PBF over NGP-based pathways (e.g., 65% CO₂ emissions in the case of HDO – BLUE_{CCS}^{M2020} pathway compared to HDO – GREEN_{CCS}^{M2020} pathway). This difference between pathways increases, even more, when the electricity mix is decarbonized (M2020 vs M2040). In the HDO – BLUE_{CCS}^{M2040} pathway, CO₂ emissions are 91% higher than in the BLUE_{CCS}^{M2040} pathway. Note that, even in DAC-based scenarios, capturing and storing atmospheric CO₂, indirect emissions incurred to meet the energy demand of this process offset these emissions, generating a positive net flow of CO₂ into the atmosphere. More information is provided in Section 3 of the Supporting information.

The second technological decision, related to the potential exploitation of LEA and the associated CO₂ emissions, leads to a pattern in the PBF metric. Scenarios entailing biogas production and combustion have a higher PBF (up to 6%) than those where LEA is directly burned. This is due to several factors, such as the fugitive emissions incurred during methane production or the lower CO₂ recovered for CCU or CCS at the end of the process (i.e., 44% lower than after direct combustion). The former aspect is particularly impactful because methane has a higher characterization factor (or global warming potential) than CO₂ on climate change. In addition, the final digestate slurry from the biogas production process is usually used as fertilizer,[46] which means that its embedded carbon will be finally released into the atmosphere [45] through biological processes.[47] Additional details about these scenarios are provided in section 3 of the Supporting information.

Regarding the CO₂ from LEA combustion, CCS scenarios related to HDO and BD achieve the best performance in all indicators except for the biogeochemical nitrogen flow, with a PBF up to 4% lower than that of diesel in the equivalent GREEN_{CCS}^{M2040} scenarios (i.e., *ceteris paribus* change). On the contrary, the absence of CCS or CCU after byproduct

cogeneration can increase impacts on climate change up to 3.7 times compared to diesel (GRAY²⁰²⁰_{NoCCU} scenario). Moreover, in GRAY or YELLOW pathways disregarding the use of LEA, impacts on climate change can reach values 3.6 times higher than BAU, thus highlighting the importance of exploiting this byproduct through CCU or CCS strategies.

Utilizing the CO₂ from cogeneration in the cultivation stage (CCU) results in higher CO₂ emissions than if that same CO₂ is stored in geological reservoirs (CCS), owing to the energy associated with CO₂ recycling and the eventual release of the CO₂ captured in the open ponds. The exception is the HTL based scenario, which achieves lower CFP and PBF in CCU scenarios than in CCS scenarios. This happens because the CO₂ generated in the HTL process covers almost entirely the CO₂ demand from the wood-microalgae mixture, which has a lower CO₂ requirement compared to 100% microalgae-based biofuel. This, in turn, translates into less CO₂ required from DAC or NGP to meet the total demand.

As shown in Figure 3, HTL scenarios achieve between 1% and 37% lower CFP than HDO scenarios when M2020 is considered. This is because the HTL process requires less energy and resources (i.e., due to the elimination of the drying and extraction process) than hydrodeoxygenation, where no extraction stage is necessary. For HTL scenarios, we consider that the CO₂ contained in HTL residue would be released into the atmosphere, in a similar way as in the digestate slurry 50. Conversely, the CO₂ in the gaseous phase can be used in both CCU and CCS processes. This CO₂ flow from HTL represents 43% of the CO₂ that can be recovered after LEA combustion. This lower CO₂ availability makes the PBF of the HTL – GREEN^{M2040}_{CCS} scenario 3% worse than for the HDO – GREEN^{M2040}_{CCS} one (4.86 vs 4.74, respectively).

Interestingly, the *ceteris paribus* change of the M2020 by a mix based on renewable sources (M2040) allows to reduce the PBF of microalgae biofuels by up to 5% for HDO – GREEN^{M2040}_{CCS}. This represents a reduction in climate change impacts of 77% compared to the equivalent scenario based on M2020. Overall, changes in the electricity mix are most noticeable for HDO – GREEN_{CCS} scenarios than for the other cases due to their higher electricity requirements stemming from a greater use of biomass from microalgae. For these scenarios, CO₂ emissions and biomass use associated with the 2020 electricity mix, result in higher impacts on climate change and nitrogen flows PBs. In the case of conventional biofuels, a change in the electricity mix would produce a very small improvement in the PBF: 0.1% for biodiesel and 1% for HDO. This happens because most emissions occur during biomass cultivation, in processes related to land-use change, fertilizer use, agricultural materials, and transport, 11,51,52 unlike the microalgae-based fuels that strongly depend on the electricity matrix. In addition, wind-powered scenarios are used here as a utopic scenario for the sake of comparison (see section 3 in the

Supporting information). According to our results, the performance of the M2040 scenarios is very close to wind-based ones. As an example, the best scenario for biofuels using the M2040 mix (HDO – GREEN_{CCS}^{M2040}) shows a PBF of only 0.6% higher than using wind. 53

3.3.3. Opportunities and limitations of biofuels from microalgae

Microalgae-based fuels present an uneven performance, depending on the particular scenario assessed. In terms of climate change, the best-performing scenario corresponds to HDO – GREEN_{CCS}^{M2040}, which only occupies 25% of the SOS, 77% less than the BAU scenario. In addition, this scenario does not transgress any of the other PB considered, and shows the lowest impacts on the PBF, the CFP and HH metrics.

Since road freight transport activities represent around 6% of the PBF of the whole economy, the value obtained for SOS of climate change by HDO – GREEN_{CCS}^{M2040} which is four times lower than BAU, reflects a change in PBF of 4% (i.e., 4.94 for BAU vs 4.74 for HDO – GREEN_{CCS}^{M2040}).

At the other end of the spectrum, we find HDO – GRAY_{NoCCU}^{M2020}, which shows a transgression of the climate change PB 3.6 times larger than the BAU scenario (See Table S29 in the Supporting information).

Despite the potential of certain biofuels to help the heavy transport sector to operate within SOS, the increase of activities for microalgae biofuel production will lead to a rise in the use of resources that are currently used by other activities. For instance, satisfying the current energy demand of the heavy-duty transport sector using the lowest impact scenario (HDO – GREEN_{CCS}^{M2040}) would require approximately 18% of the world's annual electricity consumption.^{54,55} This would require a CO₂ removal (CDR) capacity of 4.75Gt CO₂, which is less than 1% of the global CDR quota proposed for global warming mitigation.⁵⁶ The situation is not better for conventional biofuels, considering that each tkm of HDO soybean requires 0.135 m² yr.¹: 29% of the world's total cultivated area in 2019 ⁵⁷ would be needed to cover the global demand for fuel for freight road activities.

3.3.4. Relative contribution to biosphere integrity

One of the well-known drawbacks of conventional biofuels is the pressure exerted on land use, which, in turn, is a threat to biodiversity ⁵⁸. Hence, while biofuels can reduce the CO₂ emissions in transportation, which is one of the main stressors of biodiversity lost, this is typically counterbalanced by larger impacts in land use ⁵⁹. On the other hand, biofuels from algae stand out for their high yield that grants a low impact on land-system change (e.g., 50 t microalgae oil ha⁻¹ yr⁻¹ vs. 0.5 t soybean oil ha⁻¹yr⁻¹). ^{11,60,61} This

translates into lower impacts on biosphere integrity compared with, e.g., soybean scenarios: up to 4.1 times lower considering the current electricity mix scenario (HDO – GREEN $_{CCS}^{M2020}$ vs. HDO soybean) and 6.5 times lower considering the 2040 sustainable electricity mix scenario (HDO – GREEN $_{CCS}^{M2040}$), as shown in Figure 4. This difference becomes smaller for BD20, where benefits in reduced use of land ($4.7 \cdot 10^{-5} \text{ m}^2 / \text{yr tkm}$ by diesel vs. $9.6 \cdot 10^{-2} \text{ m}^2 / \text{yr tkm}$ by soybean HDO) are partially offset by the larger CO₂ emissions from diesel, which constitutes 80% of the blend.

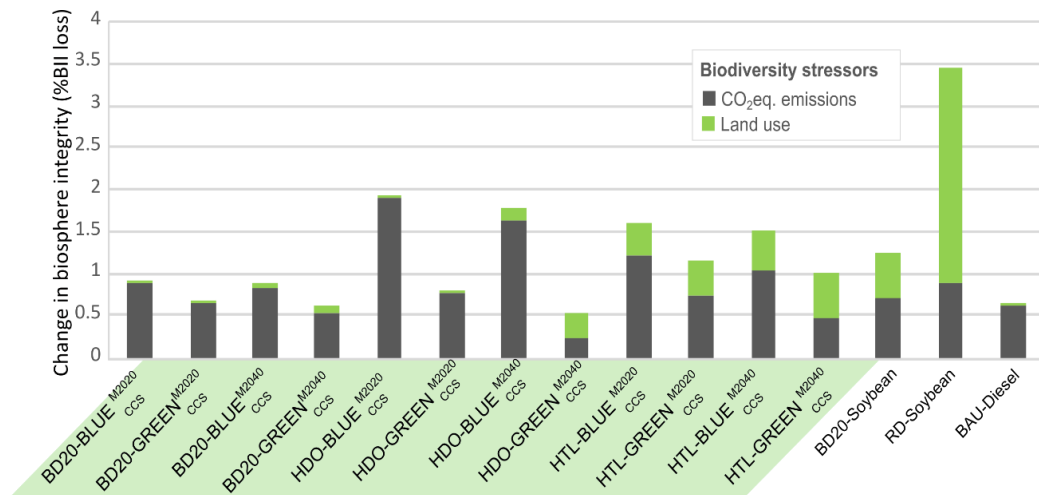


Figure 4. Impacts on change in biosphere integrity for different scenarios for the road freight activities. Expressed as the mean species abundance loss caused by the two main stressors of biodiversity loss (LU, i.e., direct land use) and CO₂ emissions.

On the other hand, Figure 4 shows that the stressors of change in biosphere integrity for the case of microalgae-based biofuels come mainly from CO₂ emissions, with very little contribution from land use. Unlike for diesel, these emissions are not due to the combustion process, but mainly stem from the energy requirements of activities such as capturing the CO₂ and recirculating water in open ponds. This is reflected by comparing equivalent scenarios with the *ceteris paribus* change of the electricity mix. As an example, the change in biosphere integrity attained by the HDO – GREEN $_{CCS}^{M2040}$ scenario is 0.5%, a 33% lower than when using the 2020 electricity mix (HDO – GREEN $_{CCS}^{M2020}$). From Figure 4, we also observe an increase in the land use stressor related to the increase in renewables for HDO – GREEN $_{CCS}^{M2020}$ scenario, contributing 59% to this impact. Renewable energies dominating the sustainable mix require a larger surface than non-renewables (up to $2.1 \cdot 10^{-3} \text{ m}^2 / \text{W}$ vs up to $5.1 \cdot 10^{-2} \text{ m}^2 / \text{W}$). 62

The only exception to this pattern is HTL, for which the land use stressor contributes between 34% and 55% of the total impact. This is due to the use of a 30% of wood pellets in the biomass mixture used with the microalgae. Although the contribution to this stressor could be reduced by modifying the share of wood in the mixture, this is the composition considered as technically and economically optimal in the literature due to the high HDO yields achieved and the current high costs of microalgae (around 590 USD/ton ash-free dry biomass).⁶³

3.3.5. Relative contribution to human health

Even though the substitution of fossil fuels with biofuels aims mostly to mitigate climate change, both types of fuels still release harmful emissions such as particulate matter (PM), non-volatile organics (NVOC), or NO_x 64,65 during combustion. So far, we have discussed the implications of these emissions for the planet, but in this section, we will turn our attention to the consequences that direct (and indirect) emissions from the heavy-duty transport sector have on HH (Figure 5). Note that these results include the emissions generated during the combustion of the three types of fuels (HDO, BD20, and diesel) in the vehicle. The details for these emissions are shown in Table S23-S25 in the Supporting information.

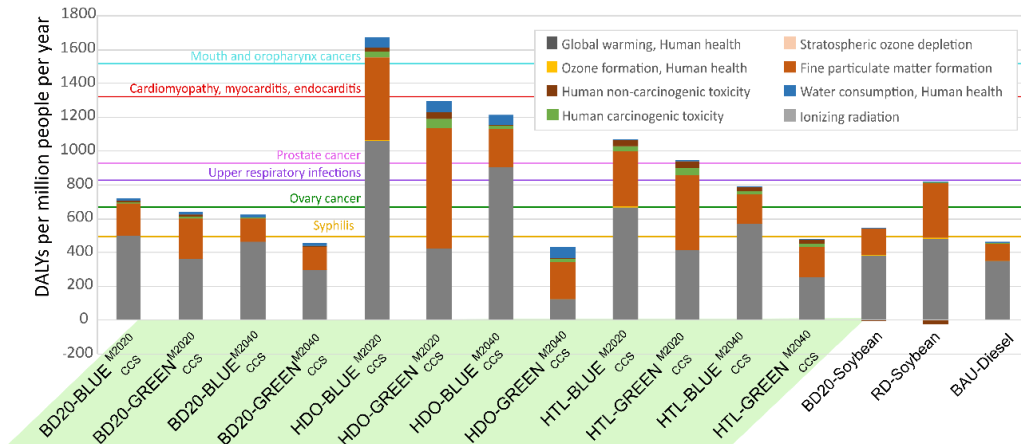


Figure 5. Impacts on human health for different scenarios for the road freight activities. Contribution of midpoints to the total impacts on human health, expressed in Disability-Adjusted Life Years (DALYs) per million people per year, based on global demand for road freight activities.

For most scenarios, the main stressors for impacts on HH are global warming and PM, with marginal contributions from water consumption and carcinogens. Some microalgae-based scenarios, such as HDO – GREEN^{M2020}_{CCS} have an impact on human health almost 3 times higher than diesel (Figure 5), despite achieving almost an equivalent PBF (Figure. 3).

This is because the emission of compounds such as PM has a different influence on some Earth's climate system than on human health, evidencing the need to carry out both analyses in parallel.

According to our study, and assuming the exclusive use of diesel (rightmost bar in Fig. 5), we estimate global emissions from the heavy-duty transport sector to be approximately 3 Gt CO₂ emissions/yr. The contribution of midpoints to the total impacts on human health from the diesel-based scenarios would cause 479 DALYs per million people per year. For the sake of comparison, it could be said that the health burden caused by heavy-duty transport sector is similar in magnitude to the one caused by syphilis.

On the other hand, microalgae-based biofuels present very different impacts depending on the scenario, ranging from 454 to 2376 DALYs per million people per year. The largest impact corresponds to the HDO – GRAY^{M2020}_{NoCCU} scenario (see Table S29). This scenario, whose performance on PBs is very similar to that of diesel, presents a three times higher impact on HH, achieving a similar magnitude to that of heart diseases such as cardiomyopathy. 44 The reason behind such a large impact is the intensive use of energy from the current fossil-based mix, which generates emissions contributing to global warming and PM formation.

Interestingly, the *ceteris paribus* change of M2020 by the mix based on renewable (i.e., M2040) sources allows to reduce the impact on HH to 454 DALYs for HDO – GREEN^{M2040}_{CCS}, i.e., 67% lower than HDO – GREEN^{M2020}_{CCS} scenario and 5% lower than diesel. This places the HDO – GREEN^{M2040}_{CCS} scenario as the one achieving the lowest impact on HH without increasing the impacts related to PBs.

3.4. Conclusions

This work compared 68 different biofuel production routes from microalgae for freight road transport under different scenarios considering a cradle-to-wheel perspective. We quantified the impact of these scenarios on seven Earth-system processes and on carbon footprint and human health to assess the potential benefits of replacing conventional fossil fuels.

Given the current state of the electricity mix and the challenges facing electric vehicles, we believe that liquid fuels will continue to play a crucial role in the transportation sector,

at least in the short to medium term, until challenges such as decarbonization of the electricity mix, battery energy densities and refueling times can be effectively addressed.

We found that conventional fossil fuels for freight road transport (e.g., diesel) are unsustainable, since they substantially transgress the climate change PB. In fact, only the freight road transport sector alone already occupies 110% of the climate change planetary boundary, leaving no place for the remaining economic activities. At the same time, alternative routes based on microalgae could substantially improve the absolute environmental sustainability level of BAU, where six scenarios based on direct air capture, carbon capture and storage, and electricity mix projected for 2040 are particularly appealing and, in principle, could operate under PBs. In contrast, when the current 2020 electricity mix is considered, HTL scenarios perform better than HDO or BD20 alternatives due to the lower use of thermal energy by avoiding the extraction stage.

Climate change boundaries show the largest share occupied by the transport sector due to their close link with fuel combustion and corresponding carbon emissions. Scenarios using fossil CO₂ for microalgae growth and the M2020 electricity mix led to an increase in the PBF and HH impacts compared with diesel in all the scenarios. On the other hand, this study shows the importance of using carbon capture utilization and storage strategies after cogeneration with lipid-extracted algae to achieve scenarios that can operate within planetary boundaries. However, even in these cases, the use of a fossil-based electricity mix can increase HH impacts relative to the BAU scenario (e.g., 2.8 times in the case of HDO – GREEN_{CCS}^{M2020}).

Finally, although several scenarios of biofuel production from microalgae fail to operate within PBs, they still achieve six times lower change in biosphere integrity compared with first-generation biofuels, such as that based on soybean. This demonstrates not only the potential of microalgae to combat climate change, but also highlights the opportunities for reducing impacts on the biosphere. The true potential of microalgae-based fuels to mitigate the impact on Earth system processes depends on their geographic location, influenced by factors such as radiation and temperature, which affect microalgae growth and yield. In this contribution, we use harmonized data from 1400 microalgae production

facilities scattered throughout the United States. This means that, the potential of microalgae to reduce environmental impacts could increase or decreased in other specific locations.

In any case, fuel production alternatives and their effects should be considered under a holistic approach to analyze the broad range of potential implications. In our study, we found that it is possible for the heavy-duty transport sector to operate within planetary boundaries while keeping impacts on HH lower than in the BAU scenario. However, algae-based fuels would require approximately 18% of the current global electricity consumption.

Promoting microalgae biofuels today as an interim solution for the transport sector could be better than using traditional biofuels, as the former can outperform the latter in terms of CO₂ emissions and changes in biosphere integrity. Combined with carbon capture and storage, these plants could remove CO₂ from the atmosphere, which is essential to achieving net-zero targets. In this context, PBs metrics, such as the one presented in this contribution, provide a robust framework for holistic assessments with the ability to minimize burden-shifting and help policymakers develop better-informed policies.

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CHAPTER IV

IV. Prospective life cycle assessment
for assessing transportation
sector alternatives.

The future of road freight transport: when and where will electric trucks be the most sustainable option?

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4.1. Introduction

According to the European Environment Agency, road transport is responsible for approximately 20% of the European Union's total greenhouse gas (GHG) emissions, with road freight transport (RFT) contributing about 25% of these [48]. RFT emissions are projected to increase by up to 22% by 2050 if current policies persist, further exacerbating the global climate crisis [49]. Despite this, ignition combustion vehicles (ICVs) will likely continue being employed in the short to medium-term. As an example, regions such as Germany, which had previously restricted the sale of ICVs after 2035, have reconsidered the regulation to secure the ongoing presence of ICVs, as long as they use low-CO₂ emissions fuels [50].

Nowadays the transport sector is in a critical transition, with many global policies striving for its gradual decarbonization through technological innovation [51,52]. One of the goals proposed by the European Commission as a means of reaching carbon neutrality by 2050 is to achieve a 40% reduction of emissions in the transportation sector by 2030 [53]. Despite this, petroleum-based fuels are still the

leaders in the transportation sector, with renewable energy sources only accounting for around 3.7% of energy supply, a 93% of which comes from biofuels [3].

In this context, the penetration in the market of low-carbon fuels such as biofuels, synthetic fuels, dimethyl ether, and electric vehicles (EVs), must be part of the integral solution to achieve the aforementioned climate objectives [54–56].

Among these options, biofuels are a promising alternative for long-distance RFT. Unlike light vehicles, which operate over short distances and whose energy demand can be met more easily, heavy-duty vehicles used for RFT require a greater amount of energy to operate and, therefore, need fuels with higher energy density. Advanced biofuels fit squarely in this application, since they exhibit a similar energy content to diesel, plus the ability to use existing engines, storage systems and infrastructures with minimal or no modifications.

On the other hand, the development and adoption of battery electric trucks (BETs) is now advancing rapidly. BETs can reach travel distances of up to 900 km and capacities of up to 14 tons of cargo [57,58]. They also have the advantage of releasing no combustion emissions during operation, and are expected to have lower operating costs than diesel trucks over their lifetime owing to lower maintenance costs [59,60]. The introduction of BETs is part of a broader trend towards the electrification of the transportation sector, where the share of electric vehicles in the global market will increase significantly in the coming years as the technology improves and becomes more cost-competitive.

Although electric trucks are frequently considered to be more sustainable than diesel trucks (dICTs), BETs are not free from life cycle emissions. As an example, life cycle CO₂ emissions of BETs can vary between 0.3 and 2.0 times those of diesel trucks [37], depending on the energy source used to charge the batteries. If the electricity comes from renewable sources such as wind or solar power, these emissions may be lower than those of dICTs, yet the use of fossil energy can lead to the opposite situation. Furthermore, in addition to indirect emissions associated with electricity production and battery manufacturing and recycling, BETs generate

significant non-exhaust emissions by braking, road wear, and tire wear, which are higher than those of ignition combustion trucks (ICTs) due to the additional weight of the batteries [61,62].

Informing decision-makers about the most sustainable vehicle technologies and fuels for the RFT sector requires a comprehensive and transparent comparison of all relevant emissions of the various options. This calls for using life cycle assessment (LCA), which allows quantification of the environmental burdens associated with the life cycle of the various alternatives, including fuel production, truck manufacturing and operation.

Various studies comparing biofuel and electric trucks have been presented to date, finding different results depending on the type of feedstock used for fuels and the primary energy sources used to generate the electricity for the batteries [37,63–65]. Sathre et al. [64] compared BETs with electricity generated from biomass, wind, and solar, with ICTs based on dimethyl ether. They found that, when based on a renewable mix, BETs have much lower GHG emissions than ICTs using diesel or dimethyl ether. Conversely, Ternel et al. [65] explored the use of biofuels, fossil fuels, and EVs on medium-size cars in 2030, finding that, in the European region, biofuel trucks have lower GHG emissions than fossil-based cars and EVs. Other works have demonstrated that the geographical region, the fuel production process, the feedstock, and the methodological assumptions employed can strongly affect the final outcome of the study [63–66].

Most of these works assume that truck technologies and the electricity mix will remain the same as today in the long term, thus disregarding the effect of improved efficiencies or the use of new decarbonization technologies. Hence, although these contributions provide valuable insights into the current environmental performance of transportation technologies, they may not capture the long-term impacts and implications of today's technological choices.

To avoid this, here, we resort to the prospective life cycle assessment (p-LCA) concept. Opposite to traditional LCA, p-LCA explicitly includes the effects of future changes in LCA datasets' background system (i.e., in all activities not explicitly

modelled). This is done by generating future life cycle inventories based on the results obtained from Integrated Assessment Models (IAMs) for various socioeconomic and environmental policies. Hence, p-LCA considers potential changes affecting environmental impacts over time, thus ensuring that long-term decisions are not made based only on today's assets.

To the best of our knowledge, few contributions have resorted to p-LCA in the context of the RFT sector [63,67]. In a pioneering work, Van den Oever et al. [63] compared two types of BETs (i.e., hybrid and 100% electric) with diesel and biofuel trucks in Europe for 2030 and 2050, finding that ICTs based on biofuels obtained through the Fischer-Tropsch synthesis could achieve lower impacts than BETs and diesel trucks.

In this contribution, we aim to assess the environmental performance of two key alternatives for decarbonizing the RFT sector over the century: ICTs powered by biofuels (bioICTs) and BETs. To accomplish this, we will perform a p-LCA of the RFT sector in different regions across the globe, utilizing a cradle-to-grave approach, i.e., covering all activities from fuel/electricity production until the end-of-life of the truck components. The comparison of the impacts incurred by these two alternatives with those stemming from the business-as-usual scenario, dominated by diesel trucks (dlCT), will allow us to quantify the reductions that could be achieved in CO₂ emissions through appropriate policies for the deployment of each of these technologies in the different regions studied. In addition, we also assess the resulting impacts on mineral resource scarcity (MRS) and on human health (HH), thus providing a comprehensive assessment of the wide implications of the deployment of these technologies in each region.

Our work extends previous research in different ways. On the one hand, we do not focus on a single zone but rather study regional variations across the globe. On the other hand, we go beyond carbon emissions and calculate impacts on two additional indicators strongly related to the transport sector. Finally, we focus on biofuels obtained from vegetable oils instead of biomass gasification, therefore exploring the potential advantages of other biofuel synthesis routes. Overall, our results can identify viable pathways towards low-carbon emissions in the RFT

sector across the globe, informing decision-makers about the benefits of supporting the most sustainable technologies according to the development level at the time.

4.2. Methodology

The present work studies the environmental performance of the RFT sector over the course of the century. The aim is to provide a comprehensive understanding of the consequences of replacing dICTs with bioICTs and/or BETs on the environment and on human health in the long term and in different regions. It is expected that the corresponding impacts will differ over time since both, technologies and policies are expected to experience changes during the century. However, LCA databases are myopic to these changes since they only provide environmental exchanges and life cycle inventories based on the current landscape. To overcome this limitation, we will introduce the following methodology, with p-LCA at its core (see Fig. 1).

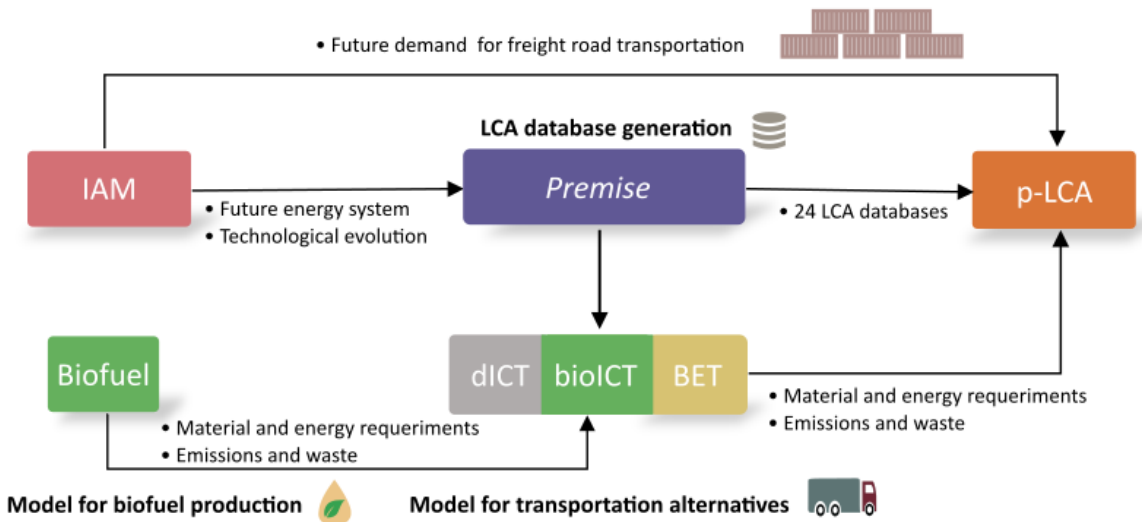


Fig. 1. Methodology overview.

The starting point is the information provided by IAMs, which forecast the potential evolution of the energy system, the sources of GHG, and the mitigation technologies used under different socio-economic narratives and climate policies. Specifically, we select two scenario projections from the REMIND IAM [68] and use the corresponding results to inform other elements of the methodology.

Next, we generate a set of LCA databases, one for each year and scenario considered, that will be used in the final p-LCA. We capitalize again on the information provided by the IAM for the selected scenarios and use the *premise* toolbox [69] to modify the Ecoinvent v3.8 LCA database accordingly [70]. The resulting databases include additional activities considering the temporal evolution of ICTs and BETs. In addition, we enter additional biofuel production activities into the databases and use them as options for the fuel consumed by bioICTs. To this end, we combine models for growing various biomass feedstocks and simulations for two fuel production processes. From these options, only the most promising alternative is kept for further analysis (see section 2.3 for further details).

With the models of the three options for the RFT sector at hand (i.e., dICTs, bioICTs, and BETs), we next perform a p-LCA of each alternative adopting a cradle-to-grave approach, i.e., considering various life cycle stages spanning from fuel production, fuel use, truck construction, and the end-of-life of the truck components. Calculations are based on meeting the annual demand for the RFT between 2020 and 2100 in the whole world, divided into 12 separate regions for consistency with the IAM employed.

Further details about the different elements of the methodology are described in the ensuing subsections.

4.2.1. Integrated assessment models

Integrated assessment models (IAMs) are crucial tools for providing insight and assessing the options and consequences of different long-term GHG emission reduction strategies [69,71]. In these models, scenarios are built by combining two essential pieces of information. On the one hand, IAMs include a series of narratives describing different future trajectories of global socio-economic conditions, i.e., the so-called shared socio-economic pathways (SSPs). On the other hand, various temperature scenarios that could result from implementing different policies [72,73], the so-called representative concentration pathways (RCPs), can also be considered. There are five socioeconomic narratives (i.e., SSP1 to SSP5), and four RCPs spanning a wide range of radiative forcing in 2100 (i.e., 2.6, 4.5, 6.0, and 8.5 W/m²) [74].

In this contribution, we capitalize on a technology-based IAM [68], REMIND, which can effectively represent how energy, economy, land, and climate systems interact and change over time. The advantage of relying on this type of model is that it can ascertain the evolution of certain technological parameters that we can use to inform later steps of our methodology. Within this IAM, we select SSP2, representing a middle-of-the-road socio-economic narrative, and concentrate on two RCPs: 3.4 and 1.9. The RCP 3.4 scenario corresponds to the continuation of current policies (CP), leading to an increase in global temperature ranging from 2 to 2.4 °C by 2100. On the other hand, RCP 1.9 aims to limit global warming to below 1.5 °C by reaching net zero around 2050 (NZ) [75], thus aligned with the aspirational goal of the Paris Agreement [76]. We will simply use CP and NZ to refer to the two scenarios considered since both rely on the same SSP.

4.2.2. Creating life cycle assessment databases for future years

The process of integrating IAM scenarios into the LCA database to represent future activities is outlined in Fig. 2. In this process, we capitalize on the *premise* v1.5.0 tool, an open-source Python library, running on BrightWay2 V.0.8.7 [31]. This tool has been designed to modify the Ecoinvent v3.8 database by incorporating IAM scenario data concerning five key energy-intensive sectors: electricity generation, cement and steel production, road freight, and passenger transport, as well as conventional and alternative fuel supplies. Moreover, *premise* includes additional activities for emerging and future technologies not originally available in the LCA database, such as hydrogen from biomass gasification, synthetic fuels, and heavy-duty trucks.[69]

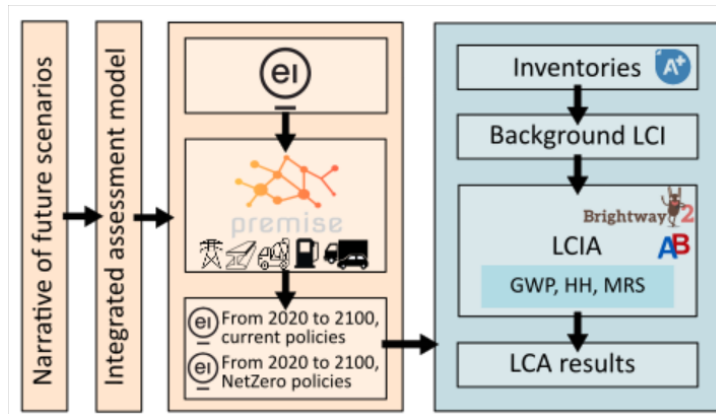


Fig. 2. Prospective life cycle assessment framework. LCI: Life cycle inventories; LCIA: Life cycle impact assessment; LCA: Life cycle assessment; GWP: Global warming potential; HH: Human health; MRS: Mineral resource scarcity.

We depart from the results obtained from REMIND under CP and NZ narratives in the 2020-2100 period. These consider changes in both technology (e.g., vehicle weight) and policies (e.g., fostering the use and development of renewable sources). We then generate new LCA databases for the period between 2020 and 2100. Databases were created at a 5-year interval until 2050 and subsequently at a 10-year interval from 2050 to 2100, thus resulting in a total of 12 databases for each scenario (i.e., 24 databases in total). This choice was made considering that most of the technological improvements considered in *premise* for the RFT sector vary only until 2050. The only exceptions are the electricity mixes of the different regions, which do vary until 2100 (Fig. 3).

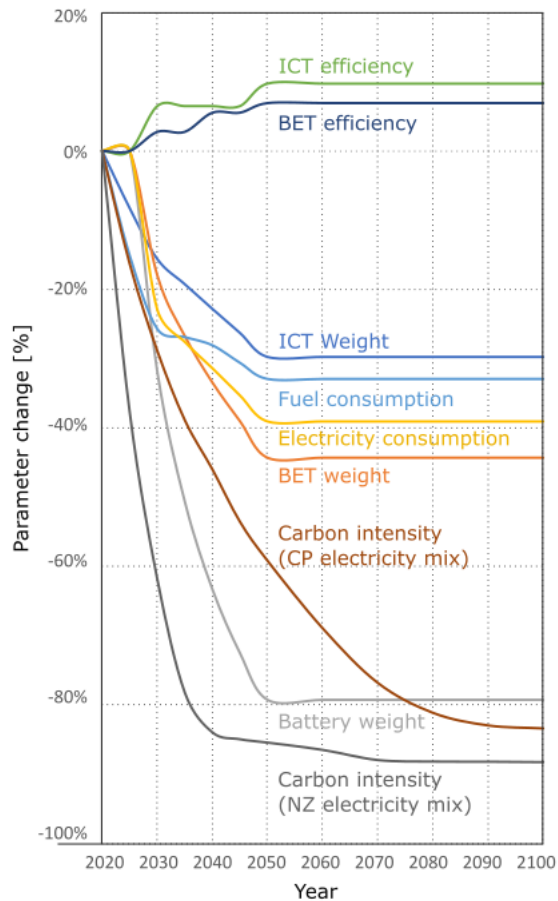


Fig. 3. Change in parameter values used to model the road freight transport sector over time. ICT efficiency: Efficiency from tank-to-wheel [%]; BET efficiency: Efficiency from battery-to-wheel [%], ICT weight: Total mass excluding driver and cargo [kg]; BET weight: Total mass excluding driver and cargo [kg]; Fuel consumption: Tank-to-wheel energy consumption of ICTs [kJ/km]; Electricity consumption: Battery-to-wheel energy consumption of BETs [kJ/km]; Battery weight: Total battery mass [kg]; Carbon intensity (CP) GHG emissions of the global electricity mix under current policies [kg CO₂ eq./MWh]; Carbon intensity (NZ): GHG emissions of the global electricity mix under current policies [kg CO₂ eq./MWh].

4.2.3. Modelling road freight transportation technologies

Models for ICTs and BETs are sourced from the LCA databases generated with *premise* [69]. This implies seven models for each truck type (i.e., one for each year considered until 2050) are available. These models are based on long-haul trucks with a capacity of 40 t, a payload of 14 t, and an autonomy of 800 km with a full tank/battery. For ICTs, the driving mass of the truck is set to 14.9 t. However, the

driving mass of BETs will vary over time because of the improvement in batteries. For some context, the battery required to achieve the same autonomy as an ICT would weight 7.8 t in 2020 but only 1.6t in 2100 [69]. As a result, the weight of BETs would vary between 22.8 t in 2020 and 16.4 t in 2100, ultimately affecting energy consumption and fugitive emissions. Information regarding both truck models is provided in Table S9 in the supplementary material.

In addition, the environmental impact of BETs is highly dependent on the composition of the electric mix used to charge their batteries. To account for this, we source the evolution of the electricity mix in the different regions from REMIND [77], and integrate this information in all the activities related to ICTs and BETs in the 24 LCA databases: from the manufacture of the truck components to the generation of the electricity for the batteries.

On the other hand, the standard model for dICTs in *premise* is used as a basis to create two alternative models for bioICTs: (i) one using a 20% blend of biodiesel (BD) with diesel, and (ii) another model fully relying on hydrotreated vegetable oil (HVO) as fuel. To this end, we assume that BD and HVO have similar characteristics to diesel. This allows us to obtain the alternative bioICT models by simply replacing an equivalent amount of diesel in the ICT models for the different years considered.

Note that both BD and HVO can be obtained from different biomass sources. Hence, to simplify subsequent analyses, we first shortlist the most promising combination of biomass feedstock and biofuel type (i.e., BD vs HVO) by means of a preliminary techno-economic and environmental assessment (TEEA) of a set of alternatives (Fig. 4). This preliminary selection process is based on global (i.e., not region-specific) LCA models for 2020 and also considered the use phase of biofuel in ICT, as will be explained next.

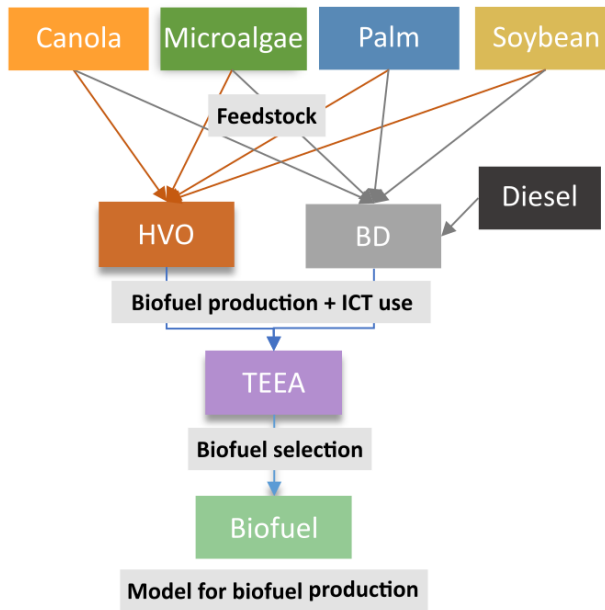


Fig. 4. Process flow diagram for the selection of the best performing biofuel. HVO: Hydrotreated vegetable oil; BD: Biodiesel; TEEA: Technoeconomic and environmental analysis.

We start by modelling LCA activities for BD and HVO production from various feedstocks. This is done by combining LCA models to produce vegetable oils from different biomass sources with models for transforming these oils into either BD through transesterification or HVO via hydrodeoxygenation.

For the former, we consider four biomass feedstocks: soybean, canola, palm, and microalgae. The first three feedstocks are selected because they are state-of-the-art sources for producing HVO [78]. In the case of microalgae, we highlight its lower land and water use requirements [79,80]. The LCA models for producing vegetable oils from soybean, canola, and palm are readily available in the Ecoinvent database, while for microalgae oil, we source the LCA data from the literature [70,80].

Then, process simulation is employed to quantify the energy and material requirements for the two routes considered for biofuel production. These simulations, outlined in Fig 5 and Fig. 6, were developed in Aspen Plus v12 based on the data from Apostolakou et al. [81] and Lee et al. [82] for BD production [81,82] and on data from Huo et al. [83] for the hydrodeoxygenation process [83].

The two simulations use triolein to mimic vegetable oil, regardless of the biomass feedstock [82,84,85]. This assumption, which allows us to perform only two (instead of eight) simulations, is underpinned by the fact that triolein is one major component of fatty acids for various vegetable oils [86,87], representing around 40–80% of their mass [88]. Both simulations were optimized to obtain the design and operating conditions yielding the minimum total annualized costs, i.e., considering both investment and operation. Energy integration (not shown in Figs. 5 and 6) was also applied to further improve the designs. Ultimately, the outputs of the simulations are used to evaluate the economic and environmental performance of these two transformation routes. Note that the environmental performance is based on LCA, where an economic allocation was used to distribute the environmental impacts among the different co-products generated during the transesterification and hydrodeoxygenation processes. Calculations for biofuel production and technoeconomic assessment are summarized in section 1 of the Supplementary material.

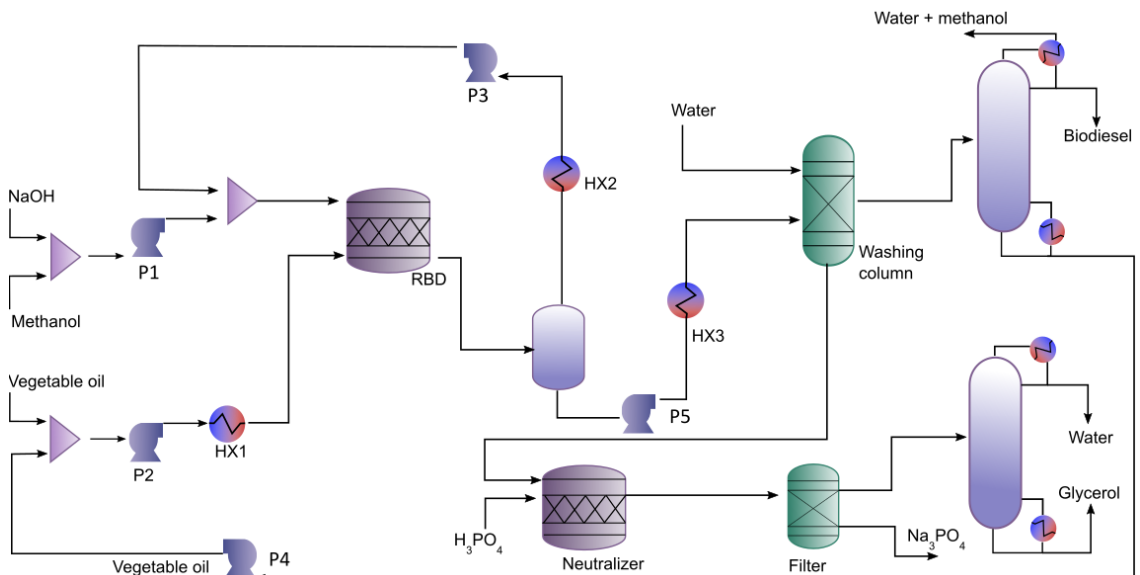


Fig. 5. Process flow diagram for biodiesel production.

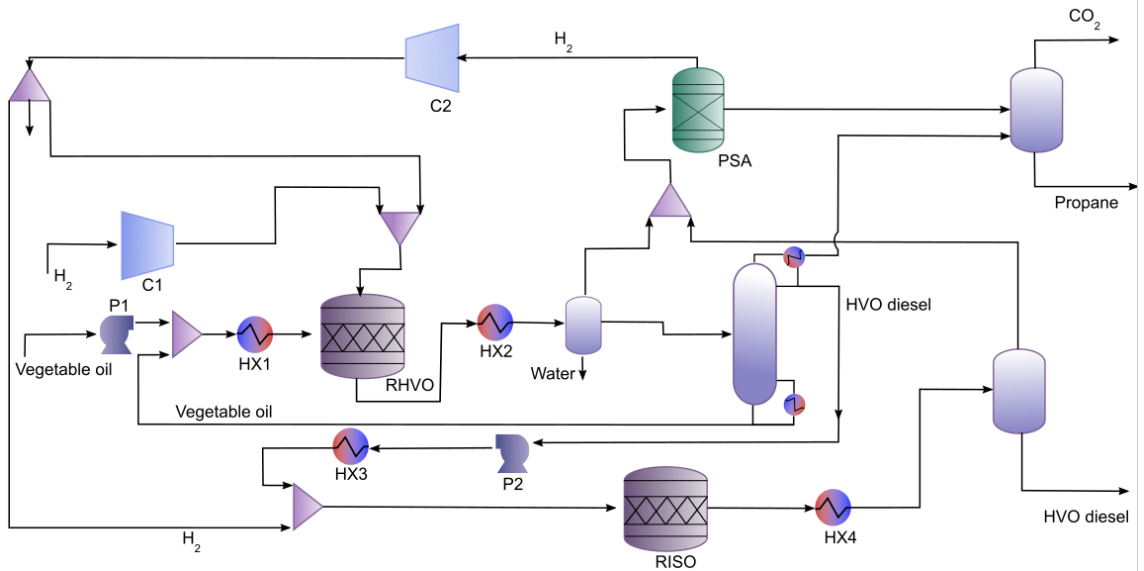


Fig. 6. Process flow diagram for hydrotreated vegetable oil production.

With the models for the eight biofuel alternatives at hand, we finally screen them based on their GWP and production costs, selecting only the best candidate for further comparison with dICTs and BETs. Results for this selection process are briefed later in section 3.1, while further details can be obtained from sections 2 and 3 of the Supplementary material. Finally, the selected alternative is used to develop bioICT models for the 12 years considered by modifying the existing dICT models from the databases created before.

4.2.4. Prospective life cycle assessment

Analogous to conventional LCA, p-LCA also follows the four-step framework outlined in ISO 14040 and 14044 standards [27,89].

In the first phase, the goal and the scope of the study are defined. The purpose of this contribution is to evaluate the impact of different alternatives for the RFT sector from cradle to grave, that is, considering impacts from activities dedicated to fuel (or energy source) production and distribution (i.e., cradle/well-to-tank), plus those incurred during the use phase in the vehicle (tank-to-wheel, i.e., combustion in the case of ICTs and electricity consumption in the case of BETs). Note that we also cover fugitive emissions from brakes/tires and downstream emissions stemming from the end-of-life alternatives for the vehicle components (i.e., to-

grave). This assessment is carried out for two possible future scenarios (i.e., CP and NZ) and 12 different years spanning from 2020 to 2100. In all the cases, the functional unit is the total demand for RFT activities expressed in kilometers traveled per ton transported per year (tkm/yr).

The second phase of the LCA identifies and quantifies the so-called life-cycle inventories (LCIs), i.e., the emissions, waste generated, and feedstock requirements associated with each RFT option p along its life cycle. For the case of dICTs and BETs, LCIs are directly retrieved for the different years from the LCA databases created with *premise*. For bioICTs, mass and energy balances from the process simulations and previous studies [24][36] are translated into the corresponding LCIs using specific activities from these databases (see Tables S7-S8). This can be seen in Eq. (1), where the LCIs associated with resource or emissions e (LCI_e , e.g., kg CO₂ emitted per tkm using a bioICT) are obtained from the net consumption of raw materials (RM_r) and utilities (UT_u) obtained from the process simulations, and the corresponding LCI entry per unit of raw material and utility ($\omega_{e,r}^{RM}$ and $\omega_{e,u}^{UT}$, respectively), as sourced from the databases. Note that some of these parameters also vary depending on the year t (e.g., $t = 2030$), the geographical location g (e.g., $g = \text{Europe}$), and the corresponding IAM scenario s (e.g., $s = \text{Net Zero policies}$) considered in each case.

$$LCI_{e,g,p,s,t} = \sum_r RM_{r,p} \omega_{e,r,g,s,t}^{RM} + \sum_u UT_{u,p} \omega_{e,u,g,s,t}^{UT} \quad \forall e, g, s, t, p = \text{bioICT} \quad (1)$$

The third phase of the LCA involves assessing the damage produced by the LCIs into different environmental categories. This can be done with the state-of-the-art impact assessment model, ReCiPe 2016 [90], which allows to calculate impacts on different midpoint and endpoint categories. Among all the options available, we focus on three categories that previous research deemed key for the transport sector: global warming potential (GWP) [91,92], mineral resource scarcity (MRS) [93–95], and human health (HH) [96,97]. The rationale behind this choice is explained next.

RFT has a substantial impact on climate change, primarily due to its reliance on fossil fuels. Incorporating the GWP indicator in our analysis allows us to quantify

the climate benefits of transitioning to more sustainable options in the RFT. BETs offer the advantage of not having exhaust gases; however, the generation of electricity used to charge batteries may introduce hazardous compounds. Including the HH indicator allows us to compare the impact of combustion gases with those associated with electricity generation and other non-exhaust emissions. Finally, it is crucial to consider the environmental challenges associated with the manufacturing, use, and disposal of batteries for BETs, which involve materials such as nickel, lithium, cobalt, copper, and rare-earth metals. To this end, MRS will be applied [93–95].

Eq. (2) is used to determine the impacts caused by each RFT alternative p on the three environmental categories b considered, in every region g , for every year t , and scenario s ($IMP_{b,g,p,s,t}$). To this end, we multiply LCIs obtained from Eq. (1) (e.g., kg CO₂ released per tkm transported) by a characterization factor ($CF_{b,e}$) translating LCIs into impacts on each category b . Characterization factors for the three indicators are based on a hierarchical perspective, which integrates environmental impacts over a 100-year time horizon. Finally, yearly volumes of load transported in each region, year and scenario ($TT_{g,s,t}$, in [tkm/yr]) are used to scale results up to the functional unit.

$$IMP_{b,g,p,s,t} = \sum_e LCI_{e,g,p,s,t} CF_{b,e} TT_{g,s,t} \quad \forall b, g, t, s, p \quad (2)$$

The yearly volumes of load transported utilized in Eq. (2) are obtained as follows. We first retrieve the energy consumption of the sector ($TTECT_{g,s,t}$, in [EJ/yr]) for the period 2020-2100 from the category labelled "Final Energy-Transport-Road" in REMIND [77]. Then, this value is divided by the current RFT energy consumption ($EC_{p,t}$, in [kJ/tkm]) of a typical dICT (see Eq. (4)). We consider a weighted average (75%-25%) [98] between the fuel consumption of a long haul with a payload of 14 t (i.e., 729 kJ/tkm for 2020), and that of a regional delivery truck with a payload of 2 t (i.e., 2989 kJ/tkm for 2020) [99]. This EC represents the total energy required per tkm.

$$TT_{g,s,t} = \frac{TTECT_{g,s,t}}{EC_t} \quad \forall b, g, t, s \quad (3)$$

With the total impacts for every region, year and alternative at hand ($IMP_{b,g,p,s,t}$), it is possible to ascertain the cumulative GWP savings ($\Delta IMP_{b,g,p,s,t}$) that could be achieved up to a certain year t , by replacing dICTs with a sustainable alternative (embedded in set SUS in Eq. (4)). Specifically, we consider three of such alternatives: (i) bioICTs, (ii) BETs, (iii) and a utopic scenario where we deploy the best of the two options in every year, region and policy. It is this last alternative that allows us to explore the potential benefits of the timely introduction of appropriate environmental policies in each region on global warming.

$$\Delta IMP_{b,g,p,s,t} = \sum_{t'} IMP_{b,g,p',s,t'} - \sum_t IMP_{b,g,p',s,t'} \quad \text{Eq.5}$$

$$\forall b, g, s, p = dICT, p' \in SUS, t, t' | t' \leq t$$

4.3. Results

4.3.1. Selection of the candidate biofuel

Fig. 7 shows the results obtained from the TEEA of the eight biofuel alternatives for bioICTs (see section 2.3). Results from BETs and dICTs are also depicted for the sake of comparison. This analysis will allow us, not only to select the most promising biofuel, but also to provide more insight into the drivers behind the performance of the different options for the RFT sector.

As a general trend, we observe that bioICTs based on BD have relatively similar performance among them, and not far away from that of diesel. This is because these options are limited by the amount of BD that can be included in the blend. As an example, bioICTs fueled with canola BD and palm BD only improve the GWP of dICT by 12% and 8%, respectively, since the emissions from the diesel in the blends already account for 48% and 64% of the total GWP of these options (Fig. 7a). In turn, the cost increase for BD bioICTs is also more modest than for other options, e.g., 13% and 8% when using canola and palm BD (Fig. 7b). The case of soybean BD is also worth mentioning, since this crop achieves worse

environmental (and economic) performance than diesel. This is primarily attributed to the farming stage, where factors such as a high land-use, machinery use, and transportation, penalize this alternative [100] .

The fact that HVOs can be used alone (i.e., without diesel) in bioICTs is translated into a more distinct performance among themselves and compared with dICT. For instance, vegetable oil production represents 84% of the total GWP for soybean, while it only accounts for 33% of the total emissions for microalgae-based HVO. From a GWP perspective, bioICTs based on microalgae HVO are the best, followed by canola and palm HVO, and with soybean emerging as the worst performing HVO. BioICTs based on microalgae HVO really stand out for their low GWP (60% lower than diesel), which is achieved by combining capture and storage of the carbon contained in the lipid-extracted algae residue, and the use of carbon from direct air capture as feedstock [80]. However, the production costs of algae HVO, 250% higher than diesel, limit the use of this alternative. On the other hand, bioICTs fueled with canola or palm HVOs demonstrate significant reductions in the GWP (30% and 14%, respectively), yet these options also present some economic challenges related to the production of vegetable oil, which can represent between 72%-86% of their total costs.

Hence, as can be seen in Fig 7c, there is no single option improving dICTs simultaneously in cost and emissions (i.e., occupying the green quadrant), which suggests that any effort to make the sector more sustainable will entail economic efforts. When balancing out advantages and disadvantages, we find that bioICTs using canola HVO and palm BD achieve the largest ratio of GWP reduction to cost increase (0.41 and 0.71, respectively). Among these two options, bioICTs based on canola HVO demonstrate a superior capacity to reduce GHG emissions and, therefore, is the option selected as the bioICT for subsequent analyses.

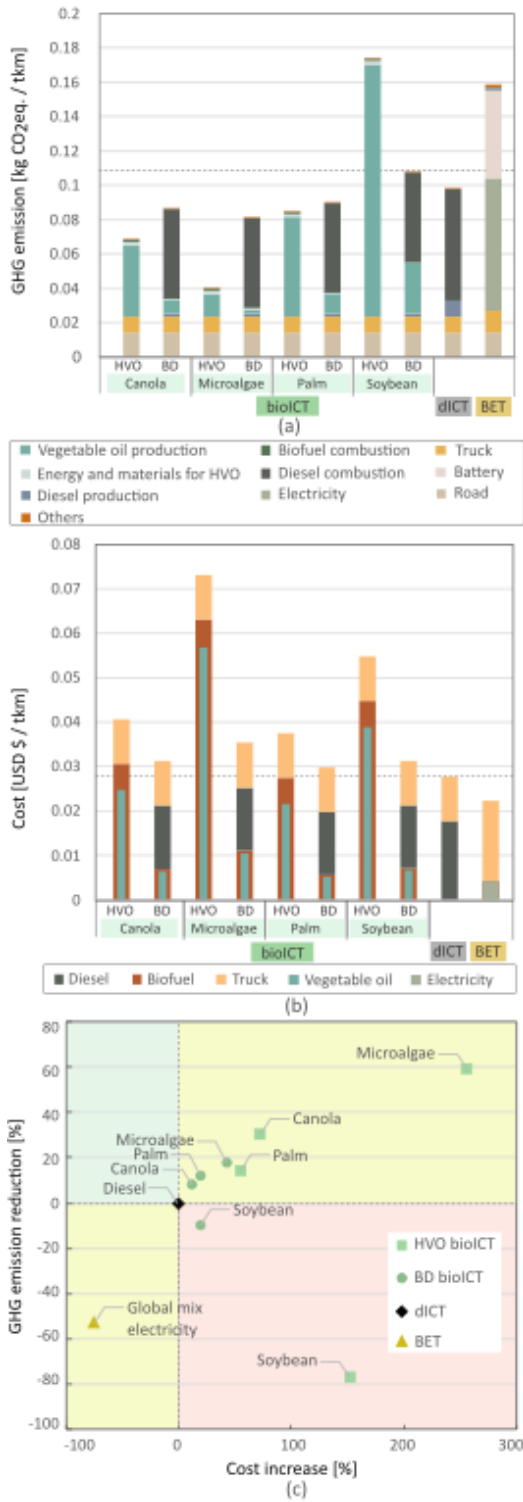


Fig. 7. Analysis of options for the road freight transport sector. (a) Breakdown of the main drivers of GHG emissions from cradle-to-grave. (b) Breakdown of the costs of the different alternatives, including fuel cost

and vehicle cost, with an internal green bar providing the cost related to the production of the vegetable oil (biofuels only). (c) GHG emissions per tkm transported (y-axis) relative to diesel, versus the cost of fuel production with respect to the cost of diesel (x-axis). GHG: greenhouse gases.

In addition, we also study the performance of BETs and compare it with that of dICTs and bioICTs. We find that, although BETs do not emit exhaust gases, their GWP is +50% larger than for dICTs in 2020. The main drivers for this are the electricity used as a power source and the production of the batteries, contributing 49% and 32%, respectively, of the total lifecycle GHG emissions for BETs (Fig. 7a). Our results evidence that electricity is 75% cheaper than the fuel cost for dICTs (Fig. 7b), yet this advantage diminishes when considering the total costs of BETs (i.e., including vehicle costs). In this case, BETs are still the most economical option for RFT, yet only 20% cheaper than dICTs, and 45% cheaper than bioICTs based on canola HVO.

The supplementary material contains further details on the techno-economic (section 1) and the environmental assessment (section 2) of all these options.

4.3.2. Evolving impacts of road freight transport under different policies and regions

Impacts from the RFT sector are expected to change dynamically over time due to the effect of variable factors like technological advancements and the evolution of electricity mixes, which, in turn, have a strong regional component. Hence, this section will explore the reduction in GHG emissions that can be achieved by satisfying the demand for RFT with bioICTs or BETs, rather than employing dICTs as mostly done today (Fig. 8). In the case of BETs, specific results are presented for the different regions studied, as significant disparities are observed between territories in variable parameters such as the electricity mix. Conversely, only a global analysis is shown for bioICTs since, in our study, most parameters affecting their performance vary in the same magnitude across regions. An exception is the carbon intensity of the electricity mixes, which does differ between regions (see Table 1). However, this variation was found to contribute marginally to GHG emissions of bioICTs (i.e., around 5%), and, therefore, it does not justify modelling regions separately. Other parameters, such as crop yields or water and soil

requirements, also expected to vary across regions, have not been considered because specific data are not available for all regions.

Table 2 Carbon intensity of electricity mix according to CP and NZ policies based on REMIND IAM

Region	Acronym	Current policies (CP) [kg CO ₂ eq/KWh]			NetZero (NZ) [kg CO ₂ eq/KWh]		
		2030	2050	2100	2030	2050	2100
Canada, Australia, and New Zealand	CAZ	147	63	63	69	60	51
China	CHA	468	263	104	121	61	64
European Union and UK region	EUR	210	82	76	113	72	56
India	IND	503	170	60	342	60	63
Japan	JAP	566	396	105	349	140	75
Latin America	LAT	173	92	136	122	85	52
Middle east	MEA	498	382	56	315	57	60
Non-EU Europe countries	NEU	182	93	62	70	56	50
Other Asian countries	OASIA	436	205	56	256	54	57
Reforming economies	REF	516	506	152	258	95	62
Sub-Saharan Africa	SSA	313	136	71	168	60	61
USA	USA	319	100	75	144	87	62

Results for the CP scenario (Fig. 8a) reveal that, during by 2030, only four regions would exhibit lower GHG emissions (10%-19%) by employing BETs instead of dICTs. These are Latin America (LAT), the European Union and United Kingdom (EU), Canada, Australia and New Zealand (CAZ), and the rest of Europe (NEU). Among them, LAT and CAZ stand out for having a high share of renewable energy in their electricity mix since 2020-2025 (i.e., 76% and 71%, respectively), with a significant contribution from hydro (58% and 48%) (see Table S11-S23 in the Supplementary material). This makes BETs a better alternative than dICTs in these regions early in the period studied.

Results also suggest that the remaining eight regions are not prepared for the wide deployment of BETs yet. Regions like China and the Middle East show electricity mixes dominated by fossil sources in 2020-2025 (65% in China and 92% in Middle East), and would need to wait until 2040-2045 before BETs show lower GWP than dICTs. By then, their electricity mix could have 57% and 30% renewable sources, respectively under CP.

Among all regions studied, reforming economies (REF) is where BETs will take the longest to surpass dICTs in terms of GHG emissions (2045-2050). This region, including countries like Russia or Uzbekistan, departs from an electricity mix composed mainly of coal (26%) and natural gas (33%). It will take until 2045-2050 to phase out coal and replace it with natural gas. This is why the improvements over BET are less significant than in other regions (42% compared to 50% less GHG emissions in 2100 under CP).

Overall, each region examined follows a different ongoing process for decarbonizing electricity generation, which results in diverse electricity mixes over time and, ultimately, in divergent timelines before BETs can improve the GWP of dICTs. This reflects the importance of coordinating efforts between the different facets of the energy sector.

The situation is very different under the NZ policy scenario (Fig. 8b), where BETs achieve lower GHG emissions than dICTs in all the regions by 2035, at latest. By then, the share of renewable energies in all regions would be at least, 52%. This translates into GHG emissions 12%-37% lower for BETs than for dICTs in all the regions: similar results to those under CP by 2050 (i.e., 15 years earlier). Hence, the early enforcement of more stringent policies for the power sector will clearly favor a quick transition towards BETs while increasing the amount of GHG emissions avoided by the substitution.

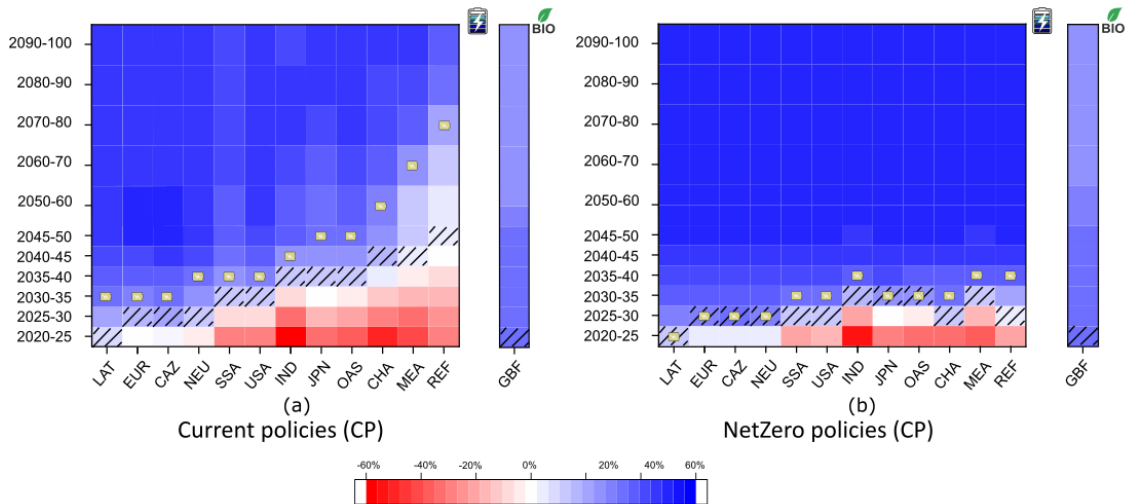


Fig. 8. Estimation of the GHG emissions reduction of road freight transport using bioICTs and BETs with respect to dICTs. The heat map illustrates the amount of GHG emission that would be avoided by replacing diesel trucks with electric or biofuel trucks under CP (subplot a) and NZ (subplot b) policies. The first twelve columns of the map depict emission reductions achieved with an electric truck (BET) for different regions. The final column, labeled "GBF," provides analogous information for biofuel trucks (bioICT) and represents worldwide results. In both cases, cells marked with lines indicate the period in which GHG emissions per tkm from BETs become lower than for dICT, while the yellow square indicates the period in which BETs have lower GHG emissions per tkm than bioICTs. GBF: Global biofuel (i.e., based on canola HBO); Region acronyms as in Table 1.

Interestingly, our results demonstrate that bioICTs will have lower GHG emissions than dICTs during the whole century, regardless of the evolution of environmental policies. An instant replacement of diesel by canola HVO, if possible, could reduce the GHG emissions from the RFT sector by 31% in 2020-2025. The fact that biofuels can use existing infrastructure for fuel distribution, as well as current truck technology, suggests that such replacement could be much faster than for BETs.

Actually, bioICTs are the best alternative for RFT in most regions during 2020-2025, emitting 29% less GHGs than BETs when considering all the regions simultaneously. This situation changes over time as electricity mixes decarbonize worldwide, and, by 2100, BETs are the best alternative by far, emitting 29% less GHGs than bioICTs and at least 45% less than dICTs. Overall, this reinforces the

idea that biofuels are a very promising interim solution as an electricity-based system for RFT is developed.

This idea is further explored in Figure 9, where we show the evolution of the annual GHG emissions for the three options considered for the RFT for the European Union and United Kingdom (EU), and China (CHA) under CP. A decreasing trend in GHG emissions is observed in both regions for all the alternatives. Although this is partially due to the decrease in the demand for RFT activities anticipated by REMIND [68], the improvement in technological parameters such as fuel consumption for ICTs, or the cleaner electricity mixes for BETs, also contribute to this trend.

According to our results for the European Union, BETs would represent a lower-GWP alternative to dICTs starting sometime before 2025, however only during the period 2025-2030 negative cumulative GHG emissions will exist with respect to dICT if CP persists. At that time, nonrenewable sources of the electricity mix are expected to represent less than 12% of electricity generation. This means that an early widespread adoption of BETs (e.g., starting in 2020) would have resulted in additional savings of 0.62 Gt CO_{2-eq} emissions compared with the continuous use of dICTs by 2035 (see bars in the bottom part of the figure). On the other hand, bioICTs always perform better than dICTs (31% better in 2020 and 28% by 2100), although BETs become the best alternative in 2035-2040 (reaching 29%-36% less GHG emissions than bioICTs in 2100). This means that the hypothetical replacement of all the dICTs in the European Union by bioICTs or BETs in 2020, would translate into 6.6 and 9.1 Gton of CO_{2-eq} saved, respectively, by the end of the century. The utopic scenario, combining the best of the two options, would allow a further increase in environmental savings up to 10.1 Gton CO_{2-eq}, only 11% larger than when using only BETs over the century. In practice, replacing the entire fleet of ICTs with BETs might take at least 20-25 years due to the long lifespan of vehicles [6,101]. Hence, these results suggest that the decarbonization foreseen in the CP of the European Union has already paved the way for the imminent adoption of BETs. In the meantime, any shift from dICTs to bioICTs would result in additional GHG emissions avoided.

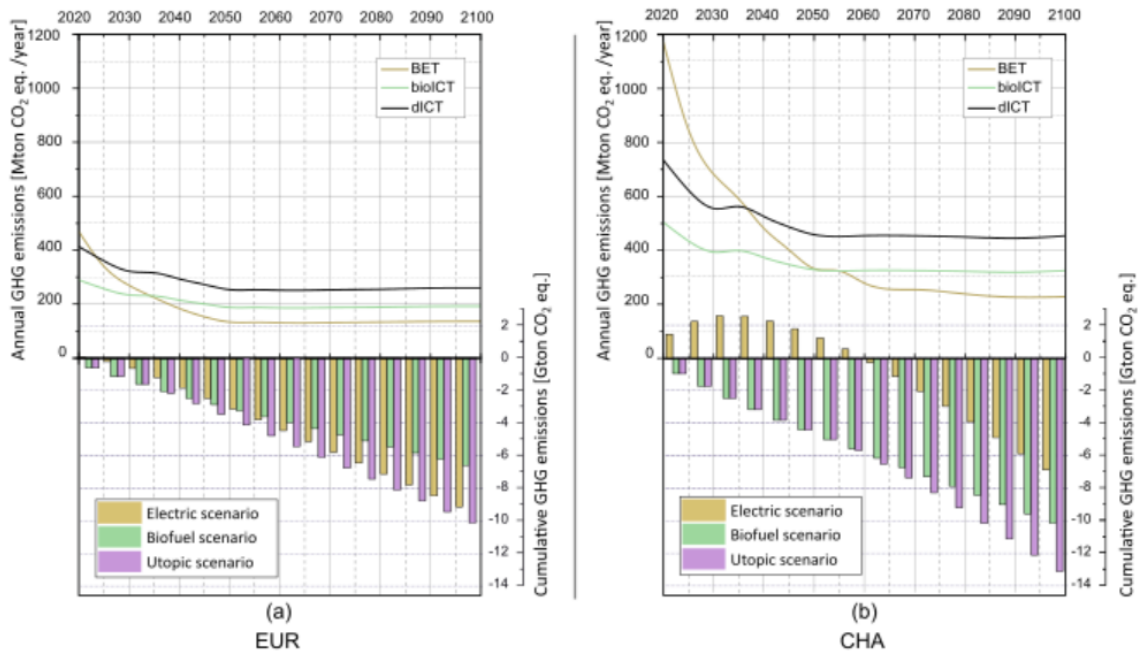


Fig. 9. Annual and cumulative GHG emissions in EUR (a) and CHA (b), under the CP scenario, using BETs, bioICTs and dICTs. Lines in the upper y-axis (left-hand side) represent the annual GHG emissions of the three alternatives. Bars in the lower y-axis (right-hand side) depict cumulative GHG emissions of each scenario, relative to those of the dICT scenario, grouped in 5-year periods.

Conversely, for China, the breakeven point between the GWP of dICTs and BETs would only arrive during 2035-2040, when renewable sources based on solar, wind, and hydro would represent around 56% of the electricity mix. This means a hypothetical replacement of all dICTs by BETs in 2020 would be counter-productive at first, resulting in larger cumulative GHG emissions early in the century that would peak during 2030-2035 at 2.8 Gt CO₂-eq. This is explained by the slow decarbonization expected for electricity under CP in China. Still, if BETs were maintained until the end of the century, 7.6Gton CO₂-eq would be saved in this region compared to the continuous use of dICTs.

The situation is even better for the bioICT scenario, where cumulative GHG emissions saved would amount to 11.3 Gton CO₂-eq by the end of the century, 33% more than with BETs. This suggests that exploiting the current infrastructure and

truck fleet through biofuels until the period where BETs have lower GHG emissions (utopic scenario) could significantly curb CO₂ emissions from the RFT sector. In the case of China, this would imply delaying the penetration of BETs until 2050-2055, reaching additional savings of 6.9 Gton CO₂-eq compared with the BETs scenario (i.e., 90% more emissions avoided), for a total of 14.6 Gton CO₂-eq avoided over the century. These savings equal to 88% of the emissions generated from 2020-2050 by dICT under CP.

Indeed, if the utopic scenario were adopted globally, emissions from the RFT sector would be 135 Gton CO₂-eq lower than with dICTs. To provide some context, this is equivalent to avoiding 0.22 °C of global temperature increase, which represents 10% of the 2 °C limit proposed by the Paris Agreement [1] (see section S1.5 in the supplementary material for calculations). This scenario is illustrated in Fig. 10, where we represent the RFT option with the lowest GHG emission for three periods (T1: 2020-2030, T2: 2030-2050, and T3: 2050-2100) in the 12 regions considered. Further results are provided in Tables S24-S35 in the supplementary material.

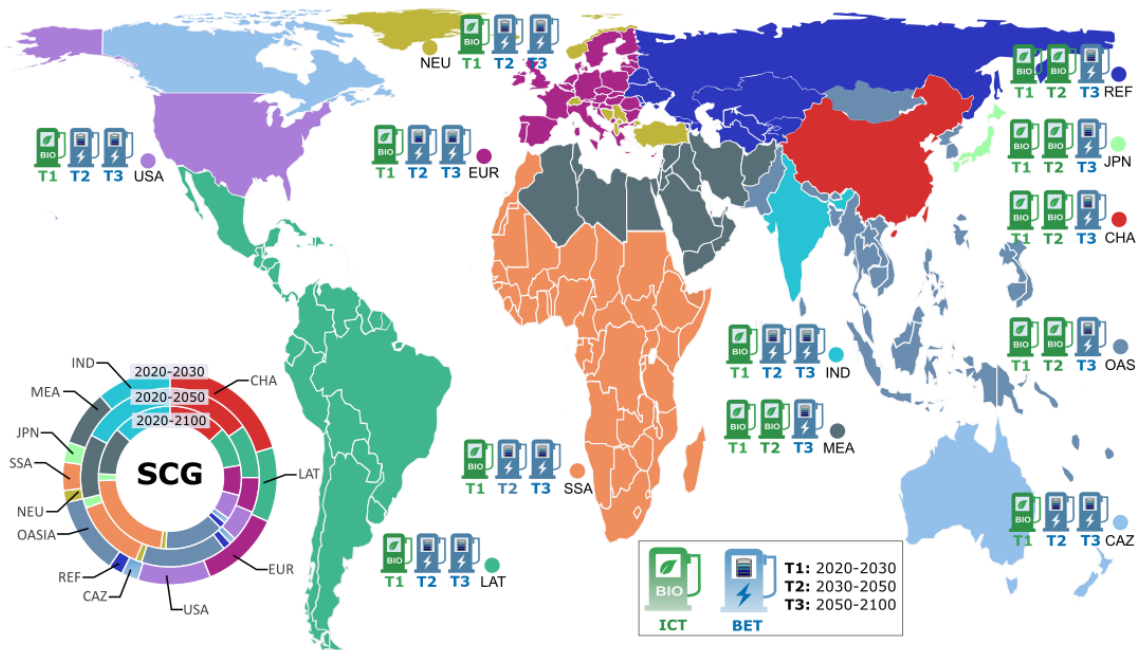


Fig. 10. Best pathway of each region for road freight transport over the century based on GWP. The icons over T1, T2, and T3 denote the freight transport alternative that achieves the lowest GHG emissions in each period (as an example, the bioICT icon for T1 shows that this alternative has the lowest GWP between 2020 and 2030). The pie chart provides the share of cumulated GHG emissions (SCG) generated by each region from 2020 to 2030 (outer circle), from 2020 to 2050 (middle circle), and from 2020 to 2100 (inner circle) under the CP scenario proposed by REMIND IAM. Region acronyms as in Table 1.

This analysis reveals that, under CP, bioICTs would be the most favorable option in all regions until 2030. From 2030 onwards, the expected increase of renewable energies over time (Tables S11 to S23 in the supplementary material) will make BETs the most favorable option in seven regions (i.e., EUR, SSA, LAT, NEU, USA, CAZ, IND). In the remaining five regions (i.e., REF, MEA, JPN, CHA, OAS), this shift will only happen after 2050.

Throughout the century (i.e., see inner circle in the pie chart in Fig. 10), certain regions such as SSA, IND, and OAS play a significant role in terms of emissions, altogether being responsible for 52% of the cumulative global emissions from the RFT sector. Population growth and increased freight activity are some of the factors affecting the share of emissions of each region. For instance, in SSA, the per-capita demand for RFT is expected to increase by 1200% along the century: from 830 tkm/person in 2020 to 10380 tkm/person in 2100. This, combined with an anticipated population surge of up to 300% by 2100 compared with 2020 [102], results in this region being responsible for 23% of the total cumulative demand for RFT, while contributing approximately 24% of the total cumulative GHG emissions during the period analyzed. This underscores the significant importance of directing efforts toward decarbonization initiatives in the SSA region.

Conversely, in the EUR, the per-capita demand for RFT is expected to rise from 9630 tkm/person to 16040 tkm/person in the same period, yet this would be combined with a slight population decline of 7% [103]. This translates into a remarkable change in the role of EUR in terms of cumulative GHG emissions (SCG): from being responsible for 12% over the period 2020-2030, to representing only 7% when 2020-2100 is considered. Moreover, ignoring the current policies

that promote the integration of bioICT and BETs [68], the use of dICTs would lead to a 68% increase in GHG emissions by the end of the century.

Within the current policy framework of the European Union, the Green Deal stands out as a key initiative. This plan aims at reducing direct GHG emissions in the road transport sector by 40% compared to 2005 levels by 2030, and by 98% by 2050 [62]. Notably, the Green Deal assumes no “direct emissions” are released from EVs owing to the absence of tailpipe emissions. Hence, considering direct emissions from dICTs and the demand for RFT for 2030 and 2050, meeting the proposed targets would require the integration of at least 26% and 88% of BETs by 2030 and 2050, respectively, in the vehicle fleet.

It is essential to recognize that although BETs show no exhaust emissions during operation, their overall life cycle still contributes to GHG emissions. Indeed, fulfilling the proposed energy demand of BETs to reach the Green Deal target would result in an increase of 143 TWh of energy demand and 38 Mton CO_{2-eq} emissions in 2030 considering the cradle-to-grave approach in the EUR. This represents 11% of the GHG emissions that would be incurred in the same region and year using only dICTs, which highlights the significance of considering life cycle approaches for policy-making.

4.3.3. Additional impacts associated with biofuel and electric trucks

Efforts to combat climate change might have undesired side effects on other aspects related to sustainable development. To address this and secure a wider view of the implications of fuel and truck substitutions, additional metrics were analyzed.

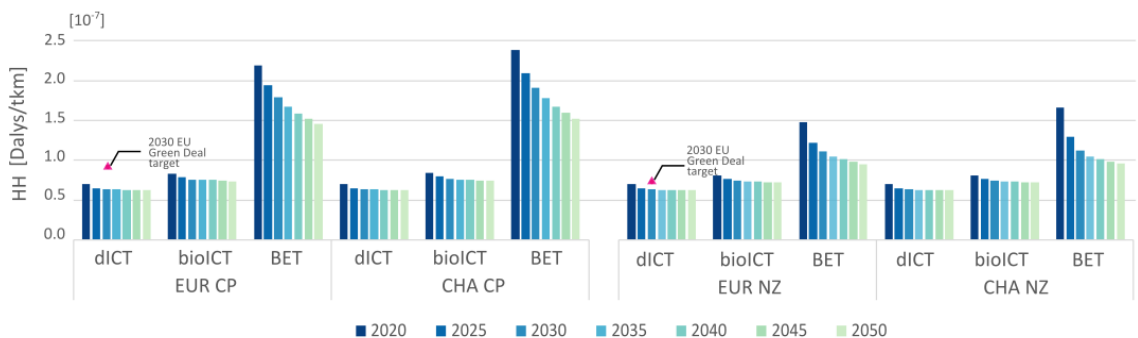


Fig. 11. Impacts on human health from freight road transport under current (CP) and NetZero (NZ) policies. EUR: European Union and UK; CHA: China; dICT: diesel-based ignition combustion truck; biofuel-based ignition combustion truck; BET: Battery electric truck. The 2030 mark in the EUR region indicates the human health impact that would result from replacing 26% of dICTs with BETs, as required to achieve the EU Green Deal target.

We begin by assessing the impacts incurred by each RFT option on human health (HH) under different policies, regions, and years (Fig. 11). We represent unitary (instead of absolute) impacts (i.e., per tkm) to prevent region size from affecting the result. We find that dICTs show the best performance among the three options considered for the two regions shown (the European Union plus United Kingdom and China) and regardless of the policy and year. These impacts remain almost constant over time, with a slight decrease of 12.3%-12.4% between 2020 and 2050 due to the increase in ICT efficiency.

BioICTs follow, showing HH impacts around 20% larger than dICT based on CP 2020 for both regions. This difference between bioICTs and dICTs is explained mainly by two factors. On the one hand, a larger amount of NO_x emissions are generated in bioICTs due to a higher combustion temperature in the presence of N₂. On the other hand, farming of the vegetable feedstock translates into higher N₂O emissions [104] and water consumption, which ultimately increases impacts on HH, too [90]. Similar to dICTs, HH impacts from bioICTs remain almost constant along the century (13%-13.4% decrease between 2020 and 2050).

Finally, we find that HH impacts from BETs are usually (i.e., for most years, regions, and policies) the largest. For the two regions explored in Fig. 11, HH impacts from BETs are between 2.1 (EUR) and 2.4 (CHA) times larger than those from dICTs under CP in 2020. In both regions, the main drivers for HH impacts are non-exhaust emissions and battery production, representing 74% of total HH impacts in EUR in 2020, and 60% in CHA for the same year. This means that replacing 26% of RFT by BETs, as needed to achieve the Green Deal target in 2030, would result in a 47% higher impact on HH than if using dICTs alone. These results evidence the need to understand the drivers behind these impacts and devote significant research efforts to prevent BETs technology from becoming a threat for human health.

For instance, non-exhaust emissions of toxic substances represent 15% of the total HH impacts from BETs in EUR under CP in 2020. These emissions include dust and other metals released from brake wear [63], as well as particulate matter resulting from abrasion caused by friction between tires and the road [105]. These impacts could be reduced by integrating new metal alloys in the brake pads, currently designed with antimony, and reducing the weight of the trucks.

The other major contributor to HH impacts from BETs is batteries. In this case, the downsizing of batteries over time translates into less resource extraction and waste generation. In addition, improvements in battery energy density result in a reduction of the truck's weight, increasing battery economy, and reducing non-exhaust emissions. This, along with the penetration of renewables in electricity mixes, was observed to reduce HH impacts during the decades by 36%: from $2.2 \cdot 10^{-7}$ DALYs/tkm in 2020 to $1.4 \cdot 10^{-7}$ DALYs/tkm in 2050 (EUR CP). In the case of EUR CP, this allows for partially closing the existing gap between BETs and dICTs: from 212% in 2020 to 133% in 2050. The situation is even better for NZ policies, where, by 2050, HH impacts from BETs will “only” be 53% larger than those from dICTs. In the case of CHA, the high reliance on coal makes HH impacts on BETs 9% higher than in EUR in 2020.

In addition, the deployment of renewable energy (e.g., photovoltaic and wind power), together with battery development, leads to an increased demand for metals [106]. To address this issue, we assess the impacts of the three options considered for RFT on metal resource scarcity (MRS) in EUR under the two policies (Fig. 12). Again, we depict unitary impacts to avoid any bias stemming from region size.

In 2020, we observe that battery materials represent 98% of the total unitary impacts on MRS when BETs are used under the CP and NZ scenarios. As aforementioned, differences in MRS impacts observed between different regions are marginal since the same truck and battery production inventories are used for all regions. Existing variations stem from varying shares of renewable sources across regions, yet these are very small since the contribution of the electricity mix is close to 2% in 2020. As an example, replacing 2020 CP's electricity mix with

that of 2050 CP in CHA would cause an increase in MRS of BETs lower than 0.2% despite the larger share of renewables.

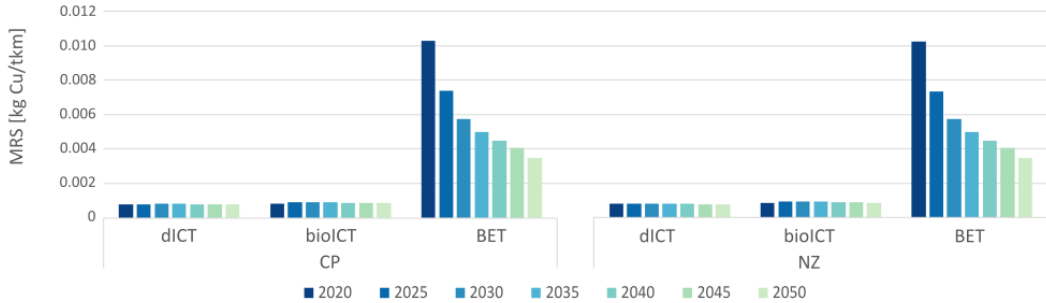


Fig. 12. Impacts on mineral resource scarcity from freight road transport under current (CP) and NetZero (NZ) policies. dICT: diesel-based ignition combustion truck; bioICT: biofuel-based ignition combustion truck; BET: Battery electric truck

On the other hand, the difference between bioICTs and dICTs is minimal (2%-11%) and stems from the use of vegetable feedstock and renewable energy. On the other hand, when BETs are compared with dICTs, MRS is 1200% higher, although this difference could decrease to 350% if they achieve measures such as increasing the energy density of the batteries, and reaching a reduction in the weight of the batteries by 79%. The penetration of renewable sources such as solar or wind, characterized by a strong reliance on metals [107], leads to increased impacts on MRS. This is especially relevant in the NZ scenario, where the more aggressive adoption of renewables causes the global electricity mix in 2050 to reach impacts on MRS 170% higher than in 2020.

4.4. Conclusions

This contribution assessed the environmental performance of two key alternatives for decarbonizing the RTF along the century: trucks powered by biofuels (bioICTs) and battery electric trucks (BETs). To this end, we divided the world into 12 regions and applied prospective LCA to each of them, utilizing a cradle-to-grave approach, i.e., covering all activities from fuel/electricity production until the end of life of the truck components. We also considered two scenarios for the evolution of

environmental policies: the continuation of current policies (CP) and rapid decarbonization through net zero strategies (NZ).

We found BETs to become the best alternative for combating climate change in the RFT sector in all regions and policies at some point between 2030-2050. This leadership would start even earlier in regions such as Latin America (2020-2025) or the European Union (2025-2030) thanks to their high share of renewables. In contrast, other regions might need to decarbonize electricity generation or reduce the energy density of batteries before adopting this technology becomes convenient. Specifically, we found that, in eight of the 12 regions considered (e.g., China or Japan), the use of BETs until 2030 would increase life-cycle GHG emissions by up to 70% compared to the continued use of current dICTs (CP).

This situation suggests that liquid fuels will continue to be an essential component for the decarbonization of the transport sector in the short to medium term, especially in regions less prepared for the early adoption of BETs. We found biofuels based on hydrotreated vegetable oil to outperform BETs in terms of GHG emissions by 11%-64% in all regions up to 2030 under CP. In addition, biofuels can leverage existing infrastructure for fuel supply, which suggests that prioritizing the early adoption of bioICTs would maximize the amount of GHG emissions avoided by the end of the century. Specifically, we found that an optimal transition from dICTs to bioICTs, and then to BETs, would translate into 134 Gt CO₂-eq avoided worldwide, and prevent 0.22°C of temperature rise. Nonetheless, it is crucial not to overlook the significance of selecting the right feedstock and biofuel type, as conventional options like biodiesel from soybean can result in emissions that are 77% higher than those from dICTs.

On the other hand, BETs are not exempt from negative side-effects. Despite having no exhaust emissions, the release of hazardous components during battery production, power generation, and fugitive emissions from brakes or tires, cause larger impacts on human health than conventional dICTs for all regions and years studied (239%-133% larger). BETs also possess higher demands for metals, with impacts on mineral resource scarcity up to 13 times larger than for ICTs in 2020 in EUR. Given the scarcity of these materials, there is an urgent need to enhance

technologies for material recovery to prevent material shortages from limiting the widespread adoption of BETs in the future.

Overall, this contribution highlights the need for comprehensive approaches to assess the sustainability level of the different options for the transport sector, addressing not only exhaust emissions, but also a myriad of other environmental problems associated with fugitive emissions, electricity generation or material scarcity. Recognizing that decarbonizing the transport sector will be a gradual process, it becomes imperative to prioritize net zero policies that accelerate the wide deployment of clean energy technologies while favoring the use of biofuels as an interim solution.

V. APPENDIX

5.1. List of publications

5.1.1. Research articles

Cabrera-Jiménez R, Mateo-Sanz JM, Gavaldà J, Jiménez L, Pozo C. Comparing biofuels through the lens of sustainability: A data envelopment analysis approach. *Appl Energy* 2021;118201. <https://doi.org/10.1016/J.APENERGY.2021.118201>.

Cabrera-Jiménez R, Tulus V, Gavaldà J, Jiménez L, Guillén-Gosálbez G, Pozo C. Microalgae Biofuel for a Heavy-Duty Transport Sector within Planetary Boundaries. *ACS Sustain Chem Eng* 2023;11:9359–71

5.1.2. Oral communications

Cabrera-Jiménez R, Tulus V, Gavaldà J, Jiménez L, Pozo C (2023). Workshop on simulation and optimization for sustainable engineering, September 2023, Santander, España

5.1.3. Book chapters

Fallanza, M; Zarca Lago, G; Tristán, C; González Lavín, G; Viar Fernández, M; Gómez Coma, L; Díaz Sainz, G. *Book of Abstracts. Workshop on Simulation and Optimization for Sustainable Engineering*. Editorial Universidad de Cantabria: Santander, 2023; pp. 53

Ferrero, L. M. M., Cabrera- Jiménez, R. C., Wheeler, J., Pozo, C., & Mele, F. D. (2022). Optimal design of sustainably efficient biorefineries supply chains. *Memorias de las JAIIO*, 8(13), 156-169.

5.2. Supplementary material

Comparing biofuels through the lens of sustainability: a Data Envelopment Analysis approach

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This document is structured in 2 parts, with section S1 providing details on data sources and data curation and section S2 presenting additional DEA results not shown in the manuscript for the sake of brevity.

S1. Data sources and data curation

The data used for the analysis of the cradle-to-wheel biofuels production were obtained using the GREET 2020 database as the main data source [108], complemented by other sources where necessary.

S1.1. Biofuel production inputs: materials and energy data

Details on the inputs (e.g., chemical reagents or energy) required to produce one liter of each biofuel, with a prior application of the economic allocation (see section 1.2), which are provided in Table S1-S9. Note that data for each biofuel type is splitted into different tables due to space limitations, with Tables S1-S5 providing data for ethanol, Tables S5 for biodiesel and Tables S7-S8 for renewable diesels. In each of these tables, two main categories can be distinguished: (i) energy required during cultivation, transport and processing of the biomass and (ii) materials used during cultivation and processing.

Table S1. Inventory flows required to produce 1 liter of ethanol as fuel – Part 1. Description of the corn processes in the Table footnote*.

	ETOH corn A	ETOH corn B	ETOH corn C	ETOH corn D	ETOH corn E
Energy (inputs)					
Diesel [MJ]	0.266	0.266	0.251	0.328	0.280
Electricity [MJ]	0.025	0.593	0.563	0.858	0.101
Gasoline [MJ]	0.079	0.080	0.075	0.098	0.084
Natural gas [MJ]	7.833	5.042	4.834	6.758	4.187
Coal [MJ]	2.944	0.432	0.414	-	0.174
LPG [MJ]	0.096	0.097	0.091	0.119	0.102
Transportation [MJ]	0.344	0.345	0.325	0.425	0.363
Residual oil [MJ]	-	-	-	-	-
Biomass [MJ]	-	-	-	-	-
Material (inputs)					
Alpha amylase [g]	0.576	0.534	0.507	0.621	0.887
Glucoamylase [g]	1.239	1.150	1.092	1.334	6.428
Cellulase [g]	-	-	-	0.882	6.428
Yeast [g]	0.629	0.584	0.553	0.211	2.051
Sulfuric acid [g]	1.066	0.989	0.938	4.536	20.854
Ammonia [g]	4.082	3.793	3.597	4.398	2.505
NaOH [g]	5.122	4.756	4.511	5.516	7.095
Corn steep liquor [g]	-	-	-	-	-
DAP [g]	-	-	-	-	-
CaO [g]	2.281	2.287	2.151	2.631	4.590
Urea [g]	-	-	-	1.464	1.252

Lime [g]	-	-	-	-	-
Fertilizer [g]	156.967	157.354	148.053	169.087	165.687
Pesticides [g]	0.470	0.471	0.443	0.506	0.496

*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; ETOH corn E: gen dry milling corn with oil extraction.

Table S2. Inventory flows required to produce 1 liter of ethanol as fuel – Part 2. Description of the corn and sorghum process in the Table footnote*.

	ETOH corn stover	ETOH corn/stover A	ETOH corn/stover B	ETOH forage sorghum	ETOH forest residue
Energy (inputs)					
Diesel [MJ]	0.692	0.282	0.282	0.528	0.466
Electricity [MJ]	-	0.101	0.814	0.079	-
Gasoline [MJ]	-	0.084	0.084	0.107	-
Natural gas [MJ]	-	0.509	5.340	1.277	0.797
Coal [MJ]	-	-	0.458	0.133	-
LPG [MJ]	-	0.102	0.102	0.164	-
Transportation [MJ]	0.072	0.366	0.366	0.478	1.329
Residual oil [MJ]	-	-	-	-	-
Biomass [MJ]	23.833	-	-	40.524	31.288
Material (inputs)					
Alpha amylase [g]	-	-	0.566	-	-
Glucoamylase [g]	-	-	1.218	-	-
Cellulase [g]	24.924	24.248	-	24.924	24.924
Yeast [g]	6.207	6.039	0.618	6.207	-

Sulfuric acid [g]	80.854	78.659	1.047	80.854	80.854
Ammonia [g]	9.703	9.439	4.017	9.703	9.703
NaOH [g]	27.491	26.744	5.037	-	27.491
Corn steep liquor [g]	30.725	-	-	-	30.725
DAP [g]	3.235	-	-	-	3.235
CaO [g]	17.788	2.404	2.404	-	17.788
Urea [g]	4.851	1.259	1.259	-	4.851
Lime [g]	-	-	-	-	-
Fertilizer [g]	52.466	145.415	145.415	213.487	-
Pesticides [g]	0.000	0.435	0.435	0.315	-

*ETOH corn/stover A: integrated corn/stover ethanol (associated with corn); ETOH corn/stover B: integrated corn/stover ethanol (associated with stover).

Table S3. Inventory flows required to produce 1 liter of ethanol as fuel – Part 3. Description of the sorghum process in the Table footnote*.

	ETOH grain sorghum	ETOH <i>Miscanthus</i>	ETOH MSW	ETOH poplar	ETOH sweet sorghum A
Energy (inputs)					
Diesel [MJ]	0.434	0.183	0.089	0.862	0.635
Electricity [MJ]	0.511	-	-	-	0.149
Gasoline [MJ]	0.225	-	-	-	0.051
Natural gas [MJ]	3.784	-	2.759	-	0.603
Coal [MJ]	-	-	-	-	0.054
LPG [MJ]	-	-	-	-	-
Transportation [MJ]	0.072	0.072	-	0.486	0.441
Residual oil [MJ]	-	-	-	-	-

Biomass [MJ]	25.090	25.647	-	27.347	28.236
Material (inputs)					
Alpha amylase [g]	0.493	-	-	-	-
Glucoamylase [g]	1.061	-	-	-	-
Cellulase [g]	-	24.924	-	24.924	7.923
Yeast [g]	0.539	6.207	-	-	5.615
Sulfuric acid [g]	0.913	80.854	80.854	80.854	25.700
Ammonia [g]	3.500	9.703	9.703	9.703	3.084
NaOH [g]	2.094	27.491	27.491	27.370	-
Corn steep liquor [g]	-	30.725	30.725	30.725	-
DAP [g]	-	3.235	3.235	3.235	-
CaO [g]	2.094	17.788	17.788	17.788	-
Urea [g]	-	4.851	4.851	4.851	-
Lime [g]	-	-	-	-	-
Fertilizer [g]	45.784	37.744	-	72.311	26.188
Pesticides [g]	1.973	0.079	-	0.202	1.135

*Sweet sorghum A: Conventional.

Table S4. Inventory flows required to produce 1 liter of ethanol as fuel – Part 4. Description of the sorghum process in the Table footnote*.

	ETOH sweet sorghum B	ETOH switchgrass	ETOH sugarcane	ETOH willow
Energy (inputs)				
Diesel [MJ]	1.055	0.225	0.405	0.620
Electricity [MJ]	0.248	0.014	0.095	0.001
Gasoline [MJ]	0.086	-	0.130	-

Natural gas [MJ]	0.635	-	0.227	-
Coal [MJ]	0.036	-	-	-
LPG [MJ]	-	-	0.199	-
Transportation [MJ]	0.732	0.072	0.356	0.488
Residual oil [MJ]	-	-	0.070	-
Biomass [MJ]	112.089	23.054	30.675	25.803
Material (inputs)				
Alpha amylase [g]	-	-	-	-
Glucoamylase [g]	-	-	-	-
Cellulase [g]	-	24.924	-	24.924
Yeast [g]	11.893	6.207	-	0.000
Sulfuric acid [g]	-	80.854	-	80.854
Ammonia [g]	-	9.703	-	9.703
NaOH [g]	-	27.370	-	27.491
Corn steep liquor [g]	-	30.725	-	30.725
DAP [g]	-	3.235	-	3.235
CaO [g]	-	17.788	-	17.788
Urea [g]	-	4.851	-	4.851
Lime [g]	-	-	9.087	-
Fertilizer [g]	43.495	44.597	82.975	8.554
Pesticides [g]	1.886	0.146	0.492	0.477

*Sweet sorghum B: Integrated.

Table S5. Inventory flows required to produce 1 liter of ethanol as fuel by gasification

	ETOH Willow G	ETOH <i>Miscanthus</i> G	ETOH Poplar G	ETOH <i>Switchgrass</i> G	ETOH Corn stover G	ETOH Forest Residue G
Energy (inputs)						
Diesel [MJ]	0.701	0.100	0.965	0.276	0.635	0.538
Electricity [MJ]	0.001	0.012	-	0.014	0.149	-
Gasoline [MJ]	-	-	-	-	-	-
Natural gas [MJ]	-	-	-	-	0.603	-
Coal [MJ]	-	-	-	-	0.054	-
LPG [MJ]	-	-	-	-	-	-
Transportation [MJ]	0.552	0.082	0.546	0.073	0.076	0.076
Residual oil [MJ]	-	-	-	-	-	-
Biomass [MJ]	30.477	32.268	22.014	24.654	23.556	36.840
Material (inputs)						
NaOH [g]	5.548	5.548	5.548	5.548	5.548	5.548
Olivine [g]	0.0007	0.0007	0.0007	0.0007	0.0007	0.0007
Synthesis catalyst [g]	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003
Tar catalyst [g]	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003
Fertilizer [g]	9.921	26.590	9.824	29.985	55.135	0
Pesticides [g]	0.051	0.089	0.227	0.148	0	0

Table S6. Inventory flows required to produce 1 liter of biodiesel.

	BD algae	BD <i>Camelina</i>	BD canola	BD corn	BD <i>Jatropha</i>	BD palm	BD tallow	BD soy
Energy (inputs)								
Diesel [MJ]	-	1.339	0.431	2.359	2.471	1.000	-	0.329
Electricity [MJ]	5.761	0.170	0.437	0.344	0.119	0.202	0.821	0.203
Gasoline [MJ]	-	-	-	0.703	-	-	-	0.070
Natural gas [MJ]	-	1.719	2.773	0.845	2.370	1.120	3.715	1.145
Coal [MJ]	-	-	-	-	-	-	-	0.229
LPG [MJ]	-	-	-	0.858	-	-	-	0.017
Transportation [MJ]	-	0.156	0.157	3.059	0.504	0.210	-	0.115
Biomass [MJ]	-	-	-	-	-	-	-	0.007
Material (inputs)								
n-hexane [MJ]	2.189	0.073	0.169	-	0.138	-	-	0.013
Methanol [kg]	0.044	0.080	0.085	0.018	0.088	0.102	1.843	0.060
Hydrogen [MJ]	-	-	-	-	-	-	0.005	0.000
NaOH [g]	0.167	0.316	0.334	0.071	0.344	0.402	0.563	0.236
CH ₃ ONa [g]	1.922	3.644	3.854	0.817	3.971	4.642	4.323	2.724
HCl [g]	0.967	1.834	1.940	0.411	1.999	2.336	2.999	1.371
H ₃ PO ₄ [g]	0.146	0.276	0.292	0.062	0.300	0.351	0.253	0.206
Citric acid [g]	-	-	-	-	-	-	0.338	-
Fertilizers* [g]	155.689	52.560	56.471	1279.635	148.607	26.797	-	12.152
Pesticides [g]	-	-	0.266	3.829	-	-	-	0.417

*The high value of fertilizers in the case of corn is due to the low percentage of oil in the corn grain (3-4%) [109]

Table S7. Inventory flows required to produce 1 liter of renewable diesel I.

	RDI algae	RDI <i>Camelina</i>	RDI canola	RDI corn	RDI <i>Jatropha</i>	RDI palm	RDI soy
Energy (inputs)							
Diesel [MJ]	-	1.281	0.396	2.623	2.344	0.907	0.319
Electricity [MJ]	6.371	0.282	0.543	0.413	0.240	0.332	0.298
Gasoline [MJ]	-	-	-	0.783	-	-	0.071
Natural gas [MJ]	-	0.815	1.769	0.722	1.351	-	0.493
Coal [MJ]	-	-	-	-	-	-	0.232
LPG [MJ]	-	-	-	0.955	-	-	0.018
Transportation [MJ]	-	0.151	0.150	3.407	0.482	0.194	0.116
Biomass [MJ]	-	-	-	-	-	-	0.007
Material (inputs)							
n-hexane [MJ]	2.419	0.071	0.163	-	0.131	-	0.014
Methanol [MJ]	-	-	-	-	-	-	-
Hydrogen [kg]	0.009	0.022	0.023	0.006	0.023	0.026	0.017
NaOH [g]	-	-	-	-	-	-	-
CH ₃ ONa [g]	-	-	-	-	-	-	-
HCl [g]	-	-	-	-	-	-	-
H ₃ PO ₄ [g]	-	-	-	-	-	-	-
Citric acid [g]	-	-	-	-	-	-	-
Fertilizers [g]	171.647	50.962	54.198	1645.320	142.060	24.849	12.298
Pesticides [g]	-	-	0.255	4.923	-	-	0.423

Table S8. Inventory flows required to produce 1 liter of renewable diesel II.

	RDII algae	RDII <i>Camelina</i>	RDII canola	RDII corn	RDII <i>Jatropha</i>	RDII palm	RDII soy
Energy (inputs)							
Diesel [MJ]	0.000	1.545	0.482	2.397	2.866	1.142	0.369
Electricity [MJ]	7.293	0.070	0.553	0.378	0.182	0.287	0.268
Gasoline [MJ]	-	-	-	0.716	-	-	0.082
Natural gas [MJ]	0.082	0.982	2.307	0.692	1.810	0.185	0.679
Coal [MJ]	-	-	-	-	-	-	0.268
LPG [MJ]	-	-	-	0.873	-	-	0.020
Transportation [MJ]	-	0.182	0.183	3.114	0.589	0.244	0.134
Biomass [MJ]	-	-	-	-	-	-	0.008
Material (inputs)							
n-hexane [MJ]	2.783	0.085	0.198	-	0.161	-	0.015
Methanol [kg]	-	-	-	-	-	-	-
Hydrogen [MJ]	0.014	0.024	0.026	0.005	0.026	0.031	0.018
NaOH [g]	-	-	-	-	-	-	-
CH ₃ ONa [g]	-	-	-	-	-	-	-
HCl [g]	-	-	-	-	-	-	-
H ₃ PO ₄ [g]	-	-	-	-	-	-	-
Citric acid [g]	-	-	-	-	-	-	-
Fertilizers [g]	197.566	61.433	65.952	1601.155	173.694	31.288	14.222
Pesticides [g]	-	-	0.310	4.791	-	-	0.488

All these values correspond to the raw inputs required for the whole manufacturing process and allocated between their different by-products (e.g., glycerin or electricity) in addition to the biofuel itself. The allocation factors used are obtained according to the methodology described in section S1.2.

S1.2. Allocation

The method used to allocate the process inputs (e.g., energy, materials and emissions loads) between the biofuel and by-products is based on the percentage of income produced by each of them [110]. The economic allocation method was adopted due to the heterogeneity of the by-products obtained in the different processes. This approach allows to standardize all products on a common basis (i.e., their economic value), regardless of the purpose of their use or their units. The economic allocation (AF_j) for each biofuel j can be obtained based on Eq. S1 as follows:

$$AF_j = \frac{MEV_j}{OEV_j} \quad \forall j \quad (S1)$$

This economic allocation factor is computed from the economic value of the main product (MEV_j) and the overall economic value of all products (OEV_j). The main product economic value (MEV_j) is calculated using the market value (MMV_j) and the amount of biofuel obtained ($Mainproduct_j$); similarly OEV_j is calculated using the amount of by-products obtained ($Byproduct_{b,j}$) and their corresponding market values ($BMV_{b,j}$).

$$MEV_j = MMV_j Mainproduct_j \quad \forall j \quad (S2)$$

$$OEV_j = MEV_j + \sum_b BMV_{b,j} Byproduct_{b,j} \quad \forall j \quad (S3)$$

The amount of by-products obtained ($Byproduct_{b,j}$) during the production process of the biofuel and their market value ($BMV_{b,j}$) are provided in Tables S9-S13. Note that we used average values for the period 2017-2019 to limit the effect of the fluctuation of product prices [108].

Table S9.Data used for the economic allocation of ethanol production – Part 1.

	[US\$]	Corn A	Corn B	Corn C	Corn D	Corn/ stover A	Corn/ stover B	Corn E
Main product								
Ethanol [L]	0.62	1	1	1	1	1	1	1
By-products								
Corn germ meal [kg]	0.22	-	-	0.15	-	-	-	-
Corn gluten feed [kg]	0.22	-	-	0.63	-	-	-	-
Corn oil [kg]	0.20	-	0.02	0.12	-	0.02	0.02	0.03
DGS [kg]	0.22	0.68	0.64	-	-	0.47	0.47	0.44
Electricity [kWh]	0.13	-	-	-	-	-	-	-
Grain sorghum [kg]	0.22	-	-	-	-	-	-	-

Table S10.Data used for the economic allocation of ethanol production – Part 2.

	[US\$]	Cellulosic	Grain sorghum	Sweet sorghum A	Sweet sorghum B	Forage sorghum	Sugar- cane
Main product							
Ethanol [L]	0.62	1	1	1	1	1	1
By-products							
Corn germ meal [kg]	0.22	-	-	-	-	-	-
Corn gluten feed [kg]	0.22	-	-	-	-	-	-
Corn oil [kg]	0.20	-	-	-	-	-	-
DGS [kg]	0.22	-	-	-	-	-	-
Electricity [kWh]	0.13	0.64	0.64	3.65	-	0.68	0.93
Grain sorghum[kg]	0.22	-	0.68	0.02	0.01	-	-

Table S11.Data used for the economic allocation of ethanol production – Part 3.

	[US\$]	Willow *G	Miscanthus *G	Poplar *G	Switchgrass *G	Corn Stover *G	Forest Residue*G
Main product							
Ethanol [L]	0.62	1	1	1	1	1	1
By-products							
Corn germ meal [kg]	0.22	-	-	-	-	-	-
Corn gluten feed [kg]	0.22	-	-	-	-	-	-
Corn oil [kg]	0.20	-	-	-	-	-	-
DGS [kg]	0.22	-	-	-	-	-	-
Electricity [kWh]	0.13	-	-	0.16	0.16	-	-
Grain sorghum[kg]	0.22	-	-	-	-	-	-

*G: Ethanol produced by gasification

Table S12.Data used for the economic allocation of biodiesel production.

Carbon source	[US\$]	Soy oil	Palm oil	Canola oil	<i>Jatropha</i> oil	<i>Camelina</i> oil	Tallow	Corn oil
Main product								
Biodiesel [L]	1.07	1	1	1	1	1	1	1
By-products								
<i>Camelina</i> meal [kg]	0.26	-	-	-	-	1.23	-	-
Canola meal [kg]	0.26	-	-	0.26	-	-	-	-
Corn meal [kg]	0.22	-	-	-	-	-	-	24.45
Electricity [kWh]	0.12	-	-	-	2.82	-	-	-
Fuel gas [kg]	0.38	-	-	-	-	-	-	-
Glycerin [kg]	0.55	0.08	0.08	0.08	0.08	0.08	0.08	0.08

Heavy oil [kg]	0.43	-	-	-	-	-	-	-
Palm kernel (PKE) [kg]	0.14	-	0.14	-	-	-	-	-
Propane [kg]	0.66	-	-	-	-	-	-	-
Soy meal [kg]	0.26	3.10	-	-	-	-	-	-

Table S13. Data used for the economic allocation of renewable diesel I production.

Carbon source	[US\$]	Soy oil	Palm oil	Canola oil	Jatropha oil	Camelina oil	Corn oil
Main product							
Renewable diesel I [L]	1.24	1	1	1	1	1	1
By-products							
<i>Camelina</i> meal [kg]	0.26	-	-	-	-	1.04	-
Canola meal [kg]	0.26	-	-	0.80	-	-	-
Corn meal [kg]	0.22	-	-	-	-	-	20.58
Electricity [kWh]	0.12	-	-	-	2.37	-	-
Fuel gas [kg]	0.38	0.19	0.19	0.19	0.19	0.19	0.19
Glycerin [kg]	0.55	-	-	-	-	-	-
Heavy oil [kg]	0.43	0.13	0.13	0.13	0.13	0.13	0.13
Palm kernel (PKE) [kg]	0.14	-	0.08	-	-	-	-
Propane [kg]	0.66	-	-	-	-	-	-
Soy meal [kg]	0.26	2.61	-	-	-	-	-

Table S14. Data used for the economic allocation of renewable diesel II production.

Carbon source	[US\$]	Soy oil	Palm oil	Canola oil	Jatropha oil	Camelina oil	Corn oil
Main product							
Renewable diesel II [L]	1.24	1	1	1	1	1	1
By-products							
<i>Camelina</i> meal [kg]	0.26	-	-	-	-	0.62	-
Canola meal [kg]	0.26	-	-	0.05	-	-	-
Corn meal [kg]	0.22	-	-	-	-	-	1.25
Electricity [kWh]	0.12	-	-	-	0.09	-	-
Fuel gas [kg]	0.38	-	-	-	-	-	-
Glycerin [kg]	0.55	-	-	-	-	-	-
Heavy oil [kg]	0.43	-	-	-	-	-	-
Palm kernel (PKE) [kg]	0.14	-	2.85	-	-	-	-
Propane [kg]	0.66	0.05	0.05	0.05	0.05	0.05	0.05
Soy meal [kg]	0.26	1.25	-	-	-	-	-

The allocation factors AF computed with Eq. S1 and data in Tables S9-S14 are finally provided in Tables S15 and S16 for the sake of completeness.

Table S15. Economic allocation factor applied to ethanol as fuel production.

Material based fuel	By-products	Allocation factor (AF)
Dry mill corn ethanol without oil extraction	Distiller grains and solubles (DGS)	0.810
Dry mill corn ethanol with oil extraction	DGS, corn oil	0.812
Wet milling corn	Corn oil, corn germ meal, corn	0.764

	gluten feed	
Combined dry and wet milling	-	1.000
Integrated Corn/Stover	DGS, corn oil	0.855
Gen dry milling corn with oil extraction	DGS, corn oil	0.860
Cellulosic material from willow	Electricity production	0.884
Cellulosic material from <i>Miscanthus</i>	Electricity production	0.884
Cellulosic material from poplar	Electricity production	0.884
Cellulosic material (Poplar gasification)	Electricity production	0.968
Cellulosic material from switchgrass	Electricity production	0.884
Cellulosic material (Switchgrass gasification)	Electricity production	0.968
Cellulosic material from corn St	Electricity production	0.884
Cellulosic material from forest	Electricity production	0.884
Grain sorghum	DGS, electricity production	0.732
Sweet sorghum (Conventional)	Grain sorghum, electricity production	0.568
Sweet sorghum (Integrated)	Grain sorghum, electricity production	0.954
Forage sorghum	Electricity production	0.876
Sugarcane	Electricity production	0.839

Table S16 Economic allocation factor applied to biodiesel and renewable diesel production.

Material based fuel	By-products	Allocation factor (AF)
Soy oil-based biodiesel	Soy meal, glycerine	0.557
Palm oil-based biodiesel	Palm kernel, glycerine	0.949
Canola oil-based biodiesel	Canola meal, glycerine	0.788
<i>Jatropha</i> oil-based biodiesel	<i>Jatropha</i> meal, glycerine, electricity production	0.812
<i>Camelina</i> oil-based biodiesel	<i>Camelina</i> meal, glycerine	0.745
Tallow based biodiesel	Glycerine	0.903
Algae oil based biodiesel I	Glycerine	0.905
Corn oil-based biodiesel	DGS, glycerine	0.167
Soy oil-based renewable diesel I	Soy meal, fuel gas, heavy oil	0.565
Palm oil-based renewable diesel I	Palm Kernel, fuel gas, heavy oil	0.882
Canola oil-based renewable diesel I	Canola meal, fuel gas, heavy oil	0.758
<i>Jatropha</i> oil-based renewable diesel I	<i>Jatropha</i> meal, fuel gas, heavy oil	0.778
<i>Camelina</i> oil-based renewable diesel I	<i>Camelina</i> meal, fuel gas heavy oil	0.724
Corn oil-based renewable diesel I	DGS, fuel gas, heavy oil.	0.186
Algae oil based renewable diesel I*	-	1.000
Soy oil-based renewable diesel II	Soy meal, propane	0.566
Palm oil-based renewable diesel II	Palm Kernel, propane	0.962
Canola oil-based renewable diesel II	Canola meal, propane	0.799
<i>Jatropha</i> oil-based renewable diesel II	<i>Jatropha</i> meal, propane	0.824
<i>Camelina</i> oil-based renewable diesel II	<i>Camelina</i> meal, propane	0.756
Corn oil-based renewable diesel II	DGS, propane	0.170
Algae oil based renewable diesel II*	-	1.000

*For renewable diesel from algae oil, by-products were not considered because the database used did not quantify by-products.

S1.3. Cost data

Table S17 shows the unitary cost data used to obtain the fuel production cost. The cost considers the material and energy inputs required during the farming of carbon sources and conversion to biofuel stages.

Table S17. Biomass yields for carbon sources used for the production of ethanol as fuel.

Energy								
Diesel [US\$/L]	0.84	[111]	Electricity [US\$/L]	0.13	[112]	Coal [US\$/mmBtu]	2.06	[112]
Gasoline [US\$/L]	0.74	[111]	Natural gas [US\$/mmBtu]	2.56	[113]	Biomass CHP [US\$/kWh]	0.15	[114]
LPP [US\$/l]	0.67	[115]						
Materials								
Alpha amylase [US\$/kg]	18.24	[32]	Glycosylase [US\$/kg]	18.24	[32]	Phosphate fertilizer [US\$/kg]	0.28	[32]
Ammonia [US\$/kg]	0.33	[116]	Herbicides (glyphosate) [US\$/kg]	4.94	[32]	Phosphoric acid [US\$/kg]	1.06	[32]
Calcium oxide [US\$/kg]	0.09	[32]	Hexane [US\$/kg]	0.38	[32]	**Synthesis catalyst [US\$/kg]	0.61	[32]
Cellulase [US\$/kg]	18.24	[32]	Hydrogen [US\$/kg]	3.15	[32]	Sodium hydroxide [US\$/kg]	0.08	[116]
Chlorohydric	0.15	[32]	Lime	0.14	[32]	Sodium	0.86	[32]

acid [US\$/kg]			[US\$/kg]			methoxide [US\$/kg]		
Citric acid [US\$/kg]	0.92	[32]	Methanol [US\$/kg]	0.26	[32]	Sulfuric acid [US\$/kg]	0.07	[32]
Olivine [US\$/kg]	0.031	[32]	Pesticides [US\$/kg]	10.56	[32]	Urea fertilizer [US\$/kg]	0.43	[116]
Diammonium phosphate [US\$/kg]	0.56	[32]	Potassium fertilizer [US\$/kg]	0.22	[32]	Yeast [US\$/kg]	2.82	[32]
Tar catalyst [US\$/kg]	0.016	[32]						
Oils								
<i>Camelina</i> [US\$/MT]	449.00	[117]	<i>Jatropha</i> oil [US\$/kg]	0.33	[118]	Canola oil [US\$/MT]	827.00	[119]
Corn oil [US\$/MT]	320.00	[119]	Palm Oil [US\$/MT]	655.00	[120]	Soybean oil [US\$/MT]	773.00	[120]

*To preserve consistency between GREET database and additional data retrieved, prices for energy were obtained for the USA.

** Synthesis catalyst price was obtained with the mass proportion of 12% molybdenum oxide (MoO₃), 2% potassium oxide (K₂O), 11% cobalt oxide (CoO), and 68% carbon support (activated carbon). The remaining weight is assumed to be sulfur from hydrogen sulfide.

S1.4. LCA data

Cradle-to-tank ReCiPe midpoint impacts of biofuel production are calculated from the inputs required in the different production stages and their corresponding unitary impacts (i.e., ecovectors, see Eq. 4 in the manuscript). The latter data, that is, impacts associated with the production of chemical reagents and the generation of energy are sourced from Ecoinvent version 3.7.1 [32] using the datasets detailed in Table S18.

Table S18. Inventory flows of the foreground processes sourced from Ecoinvent v3.7.

Flow	Process stage	Ecoinvent entry
Diesel	Farming	Diesel, burned in agricultural machinery//[GLO] market for diesel
Electricity	Farming	Electricity, medium voltage//[WECC, US only] market for electricity
Gasoline	Farming	Heat, central or small-scale, other than natural gas//[RoW] boiler
Natural gas	Farming	Heat, district or industrial, natural gas//[RoW] at industrial furnace
Coal	Farming	Heat, district or industrial, other than natural gas//[RoW] heat production, at hard coal industrial furnace
LPG	Farming	Heat, district or industrial, other than natural gas//[RoW] at industrial furnace
Transportation	Transport	Transport, freight, lorry 16-32 metric ton, EURO6//[RER] market for transport
Nitrogen	Farming	Nitrogen fertiliser, as N//[GLO] market for nitrogen fertiliser
Phosphate	Farming	Phosphate fertiliser, as P2O5//[GLO] market for phosphate fertiliser
Potassium	Farming	Potassium fertiliser, as K2O//[GLO] market for potassium fertiliser
Calcium	Farming	Lime, packed//[RoW] lime production, milled
Herbicide	Farming	Pesticide, unspecified//[GLO] market for pesticide
Insecticide	Farming	Glyphosate//[GLO] market for glyphosate
Residual Oil	Extraction	Heat, district or industrial, other than natural gas//[RoW] heavy fuel oil, at industrial furnace
Diesel	Extraction	Diesel, burned in diesel-electric generating set//[GLO] diesel,

		burned in diesel-electric generating set
Gasoline	Extraction	Heat, central or small-scale, other than natural gas//[RoW] heat production, light fuel oil, at boiler
Natural Gas	Extraction	Heat, district or industrial, natural gas//[RoW] heat production at boiler
Coal	Extraction	Heat, district or industrial, other than natural gas//[RoW]heat production at hard coal industrial furnace
LPG	Extraction	Heat, district or industrial, other than natural gas//[RoW] heat production, propane, at industrial furnace
Biomass	Extraction	Electricity, high voltage//[WECC, US only] heat and power co- generation, wood chips
Electricity	Extraction	Electricity, medium voltage//[WECC, US only] market for electricity, medium voltage
Renewable gas	Extraction	Heat, central or small-scale, natural gas//[RoW] heat production, natural gas
Methanol	Extraction	Methanol//[GLO] market for methanol
Residual oil	Conversion	Heat, district or industrial, other than natural gas//[RoW] heavy fuel oil, at industrial furnace
Diesel	Conversion	Diesel, burned in diesel-electric generating set//[GLO] diesel, burned in diesel-electric generating set
Gasoline	Conversion	Heat, central or small-scale, other than natural gas//[RoW] heat production, light fuel oil, at boiler
Natural gas	Conversion	Heat, district or industrial, natural gas//[RoW] heat production at boiler
Coal	Conversion	heat, district or industrial, other than natural gas//[RoW]heat production at hard coal industrial furnace
LPG	Conversion	heat, district or industrial, other than natural gas//[RoW] heat production, propane, at industrial furnace
Electricity	Conversion	Electricity, medium voltage//[WECC, US only] market for

		electricity, medium voltage
N-hexane	Extraction	Hexane//[GLO] market for hexane
Methanol	Conversion	Methanol//[GLO] market for methanol
Hydrogen	Conversion	Hydrogen, gaseous//[GLO] market for hydrogen, gaseous
NaOH	Conversion	Soda ash, dense//[GLO] market for soda ash, dense
CH ₃ ONa	Conversion	Sodium methoxide//[GLO] market for sodium methoxide
HCl	Conversion	Hydrochloric acid, without water, in 30% solution state//[RoW] market for hydrochloric acid
H ₃ PO ₄	Conversion	Phosphoric acid, industrial grade, without water, in 85% solution state//[GLO] market for phosphoric acid
Citric acid	Conversion	Citric acid//[GLO] market for citric acid
Alpha amylase	Conversion	Enzymes//[GLO] market for enzymes
Glucoylase	Conversion	Enzymes//[GLO] market for enzymes
Cellulase	Conversion	Enzymes//[GLO] market for enzymes
Yeast	Conversion	Fodder yeast//[GLO] market for fodder yeast
Sulfuric acid	Conversion	Sulfuric acid//[RER] market for sulfuric acid
Ammonia	Conversion	Ammonia, liquid//[RER] market for ammonia, liquid
DAP	Conversion	Nitrogen fertiliser, as N//[RoW] diammonium phosphate production
CaO	Conversion	Quicklime, milled, packed//[RoW] market for quicklime, milled, packed
Urea	Conversion	Urea, as N//[GLO] market for urea, as N
Lime	Conversion	Lime, hydrated, packed//[RoW] market for lime, hydrated, packed
Tar catalyst	Conversion	Dolomite//[RER] market for dolomite
Olivine	Conversion	Basalt//[GLO] market for basalt

For the combustion stage (i.e., tank-to-wheel), ReCiPe midpoint impacts were obtained based on the inventories provided by the GREET 2020 database [108] for combustion emissions and ReCiPe characterization impact factors (Eq. 5 in the manuscript). The resulting midpoint impacts for the combustion stage are provided in Table S19.

Table S19. ReCiPe midpoint impacts generated during the combustion of 1 liter of fuel in a light duty vehicle for the different biofuel types considered. Acronyms in Table footnote*.

Impact	E10	E85	BD20	RD	Gasoline	LS diesel
GWP [kg CO ₂ eq]	2.19	1.62	2.65	2.46	2.18	2.68
FWET [kg 1,4 DC eq]	$9.30 \cdot 10^{-5}$	$7.00 \cdot 10^{-5}$	$4.62 \cdot 10^{-3}$	$4.46 \cdot 10^{-3}$	$9.60 \cdot 10^{-5}$	$4.70 \cdot 10^{-3}$
FWEU [kg PO ₄ eq]	0	0	0	0	0	0
HT [kg 1,4 -DC eq]	$7.93 \cdot 10^{-3}$	$5.96 \cdot 10^{-3}$	$6.06 \cdot 10^{-2}$	$5.84 \cdot 10^{-2}$	$8.19 \cdot 10^{-3}$	$6.15 \cdot 10^{-2}$
PMFP [kg PM ₁₀ eq]	$2.47 \cdot 10^{-4}$	$1.85 \cdot 10^{-4}$	$3.75 \cdot 10^{-4}$	$3.58 \cdot 10^{-4}$	$2.56 \cdot 10^{-4}$	$3.81 \cdot 10^{-4}$
POFP [NMVOC eq]	$1.63 \cdot 10^{-3}$	$1.23 \cdot 10^{-3}$	$2.42 \cdot 10^{-3}$	$2.33 \cdot 10^{-3}$	$1.69 \cdot 10^{-3}$	$2.46 \cdot 10^{-3}$
TA [kg SO ₂ eq]	$4.30 \cdot 10^{-4}$	$3.17 \cdot 10^{-4}$	$6.23 \cdot 10^{-4}$	$5.86 \cdot 10^{-4}$	$4.44 \cdot 10^{-4}$	$6.36 \cdot 10^{-4}$
TE [kg 1,4 DC eq]	$6.72 \cdot 10^{-5}$	$5.05 \cdot 10^{-5}$	$2.50 \cdot 10^{-3}$	$2.41 \cdot 10^{-3}$	$6.94 \cdot 10^{-5}$	$2.54 \cdot 10^{-3}$

*GWP (global warming potential), FWET (Freshwater ecotoxicity), FWEU (Freshwater eutrophication), HT (Human ecotoxicity), PMFP (Fine particulate matter formation), POFP (Photochemical oxidant formation), TA (terrestrial acidification) and TE (terrestrial ecotoxicity); E10 (Gasoline fuel with up to 10% v/v bioethanol content), E85 (Gasoline fuel with up to 85% v/v bioethanol content), LS diesel (Low Sulphur diesel), RD (Renewable diesel), BD20 (Diesel fuel with up to 20% v/v FAME content).

Note that results in Table S17 do not distinguish between biogenic carbon and fossil carbon in the GWP emissions, and therefore total carbon dioxide emissions are shown regardless of their origin. To consider the CO₂ removed from the atmosphere during biomass growth, the amount of biogenic carbon contained in the fuel was obtained as a weight percentage (54.4% for ethanol, 76.2% for biodiesel and 84.9% for renewable diesel [32]) and discounted from the total carbon.

Table S20 provides information retrieved from the GREET database and processed to obtain the fuel consumption used to account for the emission generated by the combustion of the different fuels.

Table S20 Fuel consumption in a light-duty vehicle for the different biofuel types considered.

	E10	E85	BD20	RDII	Gasoline	LS diesel
Energy consumption [MJ/hkm]	282	264	235	235	282	235
Fuel consumption [km/L]	10.73	8.63	15.12	14.57	11.09	15.36

To consider the fuel consumption difference between light vehicles with one passenger and light vehicles with 5 passengers, it was considered that a 100 kg increase in vehicle load increases fuel consumption by 6.5% (gasoline cars) and 7.1% (diesel cars) [121]. In the case of public transport vehicles, an increase in vehicle load of up to 50% of its capacity results in fuel consumption of 5.6% [122]. Passengers are assumed to weigh 75 kg.

Table S21 collects additional information from GREET 2020 database employed during some calculations (e.g., water and land requirements).

Table S21 Physical and thermodynamic properties

	Lower Heating Value [MJ/kg]	Density [kg/m ³]
Gasoline	41.74	749.19
Low sulfur diesel	42.61	846.94
Methanol	20.09	794.10
Ethanol	26.95	789.35
Methyl ester (BD)	37.53	887.88
Renewable diesel I	43.56	748.93
Renewable diesel II	43.98	778.78
Propane	46.29	507.21
n-Hexane	44.74	654.88
Gaseous hydrogen	290 [MJ/m ³]	0.09
Natural gas	983 [MJ/m ³]	0.78
Coal	20545 [MJ/MT]	-

Table S22 provides the final list of values for the 12 indicators selected to assess the sustainability performance of each of the 60 biofuels considered. These data, assume: (i) a cradle-to-wheel perspective (i.e., including cultivation, biomass transport, fuel production and combustion stages), (ii) economic allocation between the biofuel and by-products and

(iii) one liter of biofuel is used as calculation basis for inputs and outputs in DEA. Some values of Table S22 are summarized in Table 1 in the manuscript.

Table S22. Inputs and outputs nominal data for the DEA model, expressed per liter of biofuel. Acronyms and description of the corn and sorghum process in Table footnote in the Table footnote*.

Fuel	Cost [US\$]	LO [m ²]	Water [m ³]	GWP [kg CO ₂ -Eq]	FWET [10 ⁻² kg 1,4- DCE]	FWEU [10 ⁻⁴ kg P- Eq]	HT [kg 1,4-DCE]	PMPF [10 ⁻³ kg PM10- Eq]	POFP [10 ⁻³ kg NMVOC]	TA [10 ⁻³ kg SO ₂ - Eq]	TE [10 ⁻³ kg 1,4- DCE]
BD20 algae	0.90	0.04	0.22	2.63	1.37	2.03	0.29	2.33	6.32	5.45	0.90
BD20 <i>Camelina</i>	0.78	2.28	1.06	2.52	0.96	0.40	0.14	1.60	5.34	4.42	0.78
BD20 canola	0.84	1.19	0.58	2.52	0.96	0.45	0.14	1.55	5.14	4.30	0.84
BD20 corn	0.74	3.65	1.24	2.60	1.06	0.51	0.15	1.91	6.24	4.97	0.74
BD20 <i>Jatropha</i>	0.74	1.09	0.50	2.56	1.06	0.45	0.15	1.71	5.64	4.63	0.74
BD20 palm	0.78	0.55	1.79	2.51	0.95	0.40	0.14	1.58	5.28	4.38	0.78
BD20 tallow	1.12	0.32	0.01	2.51	0.91	0.45	0.14	1.57	5.16	4.37	1.12
BD20 soy	0.82	2.22	0.14	2.50	0.92	0.40	0.14	1.53	5.09	4.28	0.82
E10 corn A	0.69	0.12	0.09	2.59	0.72	0.59	0.13	1.52	4.44	4.29	0.69
E10 corn B	0.69	0.12	0.09	2.58	0.71	0.58	0.12	1.50	4.42	4.27	0.69
E10 corn C	0.70	0.12	0.09	2.59	0.69	0.58	0.12	1.48	4.36	4.25	0.70
E10 corn D	0.71	0.15	0.15	2.70	0.90	0.67	0.15	1.78	5.09	5.23	0.71
E10 corn E	0.70	0.11	0.10	2.61	0.74	0.61	0.13	1.57	4.56	4.45	0.70
E10 corn stover	0.68	0.22	0.16	2.46	0.56	0.44	0.10	1.44	4.41	4.35	0.68
E10 corn stover*G	0.67	0.28	0.20	2.49	0.78	0.50	0.11	1.51	4.51	4.58	0.73
E10 corn/stover A	0.69	0.09	0.07	2.60	0.71	0.61	0.13	1.56	4.51	4.43	0.69
E10 corn/stover B	0.72	0.13	0.11	2.57	0.75	0.53	0.12	1.55	4.56	4.50	0.72
E10 forage sorghum	0.74	0.04	0.19	2.49	0.61	0.47	0.11	1.49	4.50	4.45	0.74
E10 forest residue	0.69	0.20	0.24	2.46	0.50	0.42	0.10	1.47	4.42	4.37	0.69
E10 forest residue*G	0.67	0.23	0.03	2.51	0.81	0.53	0.12	1.60	4.79	4.89	0.71
E10 grain sorghum	0.69	0.18	0.22	2.53	0.69	0.56	0.12	1.42	4.28	4.02	0.69

E10 <i>Miscanthus</i>	0.68	0.17	0.21	2.46	0.56	0.43	0.10	1.42	4.29	4.29	0.68
E10 <i>Miscanthus</i> *G	0.67	0.28	0.20	2.49	0.75	0.47	0.11	1.55	4.57	4.64	0.37
E10 MSW	0.69	0.01	0.00	2.51	0.61	0.45	0.10	1.54	4.40	4.71	0.69
E10 poplar	0.68	0.20	0.23	2.45	0.52	0.42	0.10	1.45	4.36	4.33	0.68
E10 poplar*G	0.67	0.24	0.14	2.50	0.74	0.49	0.11	1.55	4.60	4.67	0.35
E10 sugarcane	0.68	0.04	0.43	2.45	0.52	0.47	0.11	1.45	4.48	4.30	0.68
E10 sweet sorghum A	0.72	0.04	0.35	2.35	0.44	0.38	0.09	1.14	3.76	3.24	0.72
E10 sweet sorghum B	0.74	0.04	0.26	2.56	0.72	0.58	0.13	1.67	4.94	4.93	0.74
E10 switchgrass	0.68	0.28	0.23	2.46	0.57	0.44	0.10	1.42	4.28	4.30	0.68
E10 switchgrass*G	0.67	0.24	0.14	2.50	0.81	0.53	0.12	1.59	4.70	4.85	0.64
E10 willow	0.68	0.20	0.22	2.45	0.52	0.42	0.10	1.44	4.31	4.30	0.68
E10 willow*G	0.67	0.10	0.23	2.51	0.61	0.45	0.10	1.54	4.40	4.70	0.16
E85 corn A	0.34	0.93	0.77	2.12	2.89	2.42	0.45	2.86	7.35	6.21	0.34
E85 corn B	0.34	0.93	0.77	2.02	2.80	2.34	0.42	2.73	7.12	6.00	0.34
E85 corn C	0.40	0.94	0.74	2.31	2.84	2.49	0.46	3.02	7.74	7.53	0.40
E85 corn D	0.48	1.24	1.24	2.33	3.62	2.43	0.49	3.21	8.67	7.91	0.48
E85 corn E	0.36	0.93	0.80	2.09	2.89	2.41	0.43	2.80	7.22	5.94	0.36
E85 corn stover	0.22	1.82	1.32	0.75	1.20	0.85	0.16	1.53	5.52	4.40	0.22
E85 corn stover*G	0.14	1.92	0.24	0.75	2.93	0.12	0.22	1.71	6.01	4.96	0.48
E85 corn/stover A	0.34	0.71	0.62	2.04	2.58	2.41	0.43	2.76	6.84	5.77	0.34
E85 corn/stover B	0.57	1.06	0.89	1.78	2.90	1.67	0.39	2.63	7.24	6.34	0.57
E85 forage sorghum	0.70	0.28	1.59	1.02	1.65	1.16	0.23	1.96	6.26	5.27	0.70
E85 forest residue	0.33	7.17	0.089	0.74	0.73	0.71	0.17	1.78	5.64	4.54	0.33
E85 forest residue*G	0.12	8.36	0.18	0.76	2.38	0.14	0.30	2.33	7.63	5.8	0.23

E85 grain sorghum	0.31	1.52	1.88	1.91	2.97	2.38	0.46	2.84	7.74	6.58	0.31
E85 <i>Miscanthus</i>	0.21	1.37	1.75	0.72	1.21	0.79	0.16	1.33	4.46	3.91	0.21
E85 <i>Miscanthus</i> *G	0.13	2.36	1.59	0.71	2.77	1.14	0.21	1.35	4.64	3.66	0.49
E85 MSW	0.28	0.00	0.01	0.79	1.23	0.59	0.11	1.21	2.79	3.44	0.28
E85 poplar	0.21	1.68	1.95	0.71	0.93	0.72	0.15	1.60	5.06	4.20	0.21
E85 poplar*G	0.15	1.96	1.22	0.73	0.29	1.26	0.25	1.60	5.40	4.70	0.42
E85 sugarcane	0.25	1.30	3.66	0.85	1.07	1.23	0.28	2.03	6.95	5.32	0.25
E85 sweet sorghum A	0.56	0.34	2.96	0.96	1.46	1.41	0.29	2.11	6.85	5.30	0.56
E85 sweet sorghum B	0.75	0.29	2.21	1.35	2.34	1.86	0.35	2.78	8.43	6.95	0.75
E85 switchgrass	0.26	2.33	1.98	0.72	1.30	0.84	0.16	1.35	4.38	3.93	0.26
E85 switchgrass*G	0.29	2.37	1.73	0.81	2.84	1.49	0.31	2.33	7.28	6.25	0.211
E85 willow	0.25	1.68	1.90	0.68	0.86	0.68	0.14	1.48	4.70	3.95	0.25
E85 willow*G	0.28	1.96	1.21	0.69	2.43	1.28	0.27	1.90	6.46	4.87	0.184
RDI algae	1.17	0.20	0.23	1.10	2.69	7.29	0.75	4.10	8.21	6.15	1.17
RDI <i>Camelina</i>	0.61	11.04	3.97	0.58	0.80	0.44	0.10	1.04	4.00	1.82	0.61
RDI canola	0.90	5.69	2.09	0.59	0.77	0.64	0.11	0.84	3.17	1.34	0.90
RDI corn	0.42	9.05	0.50	1.01	1.47	0.99	0.19	2.58	8.53	4.53	0.42
RDI <i>Jatropha</i>	0.41	5.16	1.88	0.74	1.20	0.65	0.14	1.48	5.18	2.64	0.41
RDI palm	0.62	2.53	5.54	0.52	0.72	0.45	0.10	0.95	3.67	1.63	0.62
RDI soy	0.79	9.47	2.51	0.51	0.66	0.44	0.10	0.74	3.02	1.28	0.79
RDII algae	1.30	0.22	0.26	0.92	3.02	8.66	0.88	4.84	9.39	7.29	1.30
RDII <i>Camelina</i>	0.68	11.67	5.43	0.30	0.86	0.45	0.11	1.18	4.40	2.10	0.68
RDII canola	1.03	6.07	2.96	0.31	0.83	0.70	0.12	0.92	3.37	1.50	1.03
RDII corn	0.47	9.82	0.50	0.64	1.42	0.94	0.18	2.47	8.22	4.34	0.47
RDII <i>Jatropha</i>	0.46	5.54	2.53	0.51	1.38	0.71	0.16	1.74	5.94	3.17	0.46

RDII palm	0.70	2.79	9.02	0.23	0.77	0.45	0.10	1.07	4.05	1.91	0.70
RDII soy	0.91	11.38	3.49	0.28	0.75	0.50	0.12	0.86	3.27	1.53	0.91

*Corn A: dry mill corn without oil extraction; Corn B: dry mill corn with oil extraction; Corn C: wet milling corn; Corn D: combined dry and wet milling corn; Corn/stover A: integrated corn/stover ethanol (associated with corn); Corn/stover B: integrated corn/stover ethanol (associated with stover); Corn E: generic dry milling corn with oil extraction; GWP (global warming potential), FWET (Freshwater ecotoxicity), FWAC (Freshwater acidification), HT (Human ecotoxicity), PMFP (Fine particulate matter formation), POFP (Photochemical oxidant formation), TA (terrestrial acidification) and TE (terrestrial ecotoxicity), E10 (Gasoline fuel with up to 10% v/v bioethanol content), E85 (Gasoline fuel with up to 85% v/v bioethanol content), LS diesel (Low Sulphur diesel), RD (Renewable diesel), BD20 (Diesel fuel with up to 20% v/v FAME content). *G: Ethanol produced by gasification

S1.5. Water and land requirements

In this section, we provide the data for the calculation of water and land requirements for growing the amount of crop necessary to produce 1 liter of the corresponding biofuel. In the case of the water use indicator (*Water*), two contributions are considered: irrigation during one year (*Water^{Crops}*, retrieved from Tables S23 and S24) and the life-cycle water consumption for chemicals and energy production (*Water^{Inputs}*). The latter is retrieved from the LCI labelled as “water depletion” in Ecoinvent v3.7.

Table S23. Water requirements for growing feedstock required to produce ethanol as fuel.

Carbon source	Water requirements	Ref.
Corn [mm/year]	800	[123]
Corn stover [mm/year]	120	[124]
Forage sorghum [mm/year]	490	[125]
Forest residue [mm/year]	228	[126]
Grain sorghum [mm/year]	650	[127]
<i>Miscanthus</i> [mm/year]	700	[128]
Poplar [mm/year]	590	[129]
Sugarcane [mm/year]	2500	[130]
Sweet sorghum [mm/year]	490	[125]
Switchgrass [mm/year]	704	[131]
Willow [mm/year]	620	[129]

Table S24. Water requirements for growing feedstock required to produce biodiesel and renewable diesel.

Carbon source	Water requirements	Ref.
Algae [L/L algae oil]	242	[132]
<i>Camelina</i> [mm/year]	720	[133]
Canola [mm/year]	700	[134]
Corn [mm/year]	800	[123]
<i>Jatropha</i> [mm/year]	780	[135]
Palm [mm/year]	3500	[136]
Soy [mm/year]	700	[123]
Tallow [L/kg tallow]	13	[137]

In the case of tallow production, a water requirement of 154.5 L/kg beef [137], with a composition of 0.20 kg tallow/kg beef [138] and an economic allocation factor for tallow of 1.73% [139] were considered obtaining a water requirement of 13.4 L water/kg tallow.

The biomass yield that can be obtained over a year by adopting irrigation rates shown in Tables S23 and S24 are provided in Tables S25 and S26.

Table S25. Biomass yields for carbon sources used for the production of ethanol as fuel.

Carbon source	Biomass yield	Ref.
Corn [t/ha/yr]	17.31	[140]
Corn stover [t/ha/yr]	12.80	[140]
Forage sorghum [t/ha/yr]	94.00	[108]
Forest residue [t/ha/yr]	3.24	[35]
Grain sorghum [t/ha/yr]	9.73	[108]
<i>Miscanthus</i> [t/ha/yr]	17.00	[32]
Poplar [t/ha/yr]	13.50	[36]
Sugarcane [t/ha/yr]	67.00	[108]
Switchgrass [t/ha/yr]	10.00	[140]
Sweet sorghum [t/ha/yr]	94.00	[108]
Willow [t/ha/yr]	13.84	[36]

Table S26. Biomass yields for carbon sources used for biodiesel and renewable diesel production.

Carbon source	Oil yield	Ref.
Algae [kg oil/ha/year]	50717	[37]
<i>Camelina</i> [kg oil/ha/year]	561	[38]
Canola [kg oil/ha/year]	1140	[39]
Corn [kg oil/ha/year]	74	[140]
<i>Jatropha</i> [kg oil/ha/year]	1300	[135]
Palm [kg oil/ha/year]	3000	[136]
Soy [kg oil/ha/year]	430	[140]
Tallow [kg tallow/ha/year]	5922	[137]

In the case of tallow, the calculation was made based on a requirement of 2.2 ha/ton of beef [138], also taking into account the economic allocation factor of beef (1.73%) [139] and the composition of tallow in the beef (0.20 kg tallow/kg beef) [138].

Finally, Tables S27 and S28 show the land and water requirements as a function of the amount of fuel produced. The land requirements are calculated using the transformation yield during the conversion of feedstock to fuel (Table S27-S28) and the yield of the carbon source (Table S25-26), resulting in the area needed during one year to obtain 1 liter of biofuel. Similarly, to obtain the irrigation water requirements, the transformation yield during feedstock to fuel conversion, the yield of the carbon source and the water

requirements (Table S23-S24) are processed. Densities from Table S21 are employed where necessary.

Table S27. Land and water requirements for carbon sources to produce 1 liter of ethanol. Description of the corn and sorghum process in the Table footnote*.

Material based fuel	Transformation yield [dry kg biomass/L fuel]	Land requirement [m ² /L fuel]	Water requirements [L H ₂ O/L fuel]
Corn A ethanol	2.35	1.36	1084.30
Corn B ethanol	2.35	1.36	1084.30
Corn C ethanol	2.51	1.45	1161.46
Corn D ethanol	2.51	1.45	1161.46
Corn E ethanol	1.68	1.27	1161.46
Corn stover ethanol	3.11	2.43	291.43
Corn stover ethanol G	2.88	2.25	270
Corn/stover A ethanol	1.67	0.97	775.27
Corn/stover B ethanol	2.51	1.45	1161.46
Forage sorghum ethanol	14.84	1.58	773.63
Forest residue ethanol	3.11	9.59	2187.05
Forest residue ethanol G	3.18	9.83	2242.45
Grain sorghum ethanol	2.38	2.45	1595.45
<i>Miscanthus</i> ethanol	3.11	1.83	1279.73
<i>Miscanthus</i> ethanol G	2.88	1.70	1188.2
Poplar ethanol	3.11	2.25	1324.90
Poplar ethanol G	3.18	2.30	1358.47
Sugarcane ethanol	12.34	1.84	4606.15
Sweet sorghum A ethanol	6.625	0.70	345.35
Sweet sorghum B ethanol	14.04	1.49	732.09
Switchgrass ethanol	3.11	3.11	2187.97
Switchgrass ethanol G	2.88	2.88	2032.54
Willow ethanol	3.11	2.25	1392.27
Willow ethanol G	3.18	2.30	1427.54

*Corn A: dry mill corn without oil extraction; Corn B: dry mill corn with oil extraction; Corn C: wet milling corn; Corn D: combined dry and wet milling corn; Corn/stover A: integrated corn/stover ethanol (associated with corn); Corn/stover B: integrated corn/stover ethanol (associated with stover); Corn E: gen dry milling corn with oil extraction; Sweet sorghum A: conventional; Sweet sorghum B: integrated.

Table S28. Land and water requirements for carbon sources to produce 1 liter of biodiesel, renewable diesel I and renewable diesel II as fuel.

Material based fuel	Transformation yield	Land requirement	Water requirements
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	[kg fuel/-kg oil]	[m ² /L fuel]	[L H ₂ O/-L fuel]
<i>Camelina</i> oil-based biodiesel	1.03	1.53	1425.56
<i>Camelina</i> oil-based renewable diesel I	1.04	1.52	5486.5
<i>Camelina</i> oil-based renewable diesel II	0.9	1.54	7179.32
Canola oil-based biodiesel	1.03	0.75	736.36
Canola oil-based renewable diesel I	1.04	0.75	2759.34
Canola oil-based renewable diesel II	0.9	0.76	3702.43
Corn oil-based biodiesel	1.03	5124	7428.17
Corn oil-based renewable diesel I	1.04	5112	2698.22
Corn oil-based renewable diesel II	0.9	5177	2940.14
<i>Jatropha</i> oil-based biodiesel	1.03	0.66	609.01
<i>Jatropha</i> oil-based renewable diesel I	1.04	0.66	2413.68
<i>Jatropha</i> oil-based renewable diesel II	0.9	0.66	3072.33
Palm oil-based biodiesel	1.03	0.29	1879.3
Palm oil-based renewable diesel I	1.04	0.28	6279.23
Palm oil-based renewable diesel II	0.9	0.29	9379.33
Soy oil-based biodiesel	1.03	1.99	1217.51
Soy oil-based renewable diesel I	1.04	1.67	4440.98
Soy oil-based renewable diesel II	0.9	2.01	6172.07
Tallow based biodiesel	1.1	0.03	13.79

2. Additional results

S2.1. Complete DEA results

A non-oriented SBM approach was selected to determine the operational and environmental efficiency of 72 fuels based on renewable feedstocks and complemented with a super-efficiency SBM analysis. Efficiency and super-efficiency models were coded in GAMS v32.1 software [40]. The complete list of (super)efficiency scores obtained are presented in Table S29, while the improvement targets for inefficient units are presented as a percentage of original inputs and outputs in Table S30. A summary of these data is provided in Figs. 5 and 7 in the manuscript, respectively.

Table S29. (Super)efficiency scores obtained by the different biofuels. Description of the corn and sorghum process in the Table footnote*.

Fuel	Efficiency	Fuel	Efficiency
BD20 algae	0.749978	E10 <i>Miscanthus G</i>	1.003567
BD20 <i>Camelina</i>	1.002226	E10 MSW	1.000032
BD20 canola	1.006525	E10 poplar	1.000042
BD20 corn	0.824471	E10 poplar G	0.7701
BD20 <i>Jatropha</i>	1.015603	E10 sugar cane	1.00164
BD20 palm	1.045031	E10 sweet sorghum A	1.130111
BD20 tallow	1.163293	E10 sweet sorghum B	0.672252
BD20 soy	1.040191	E10 switchgrass	0.820161
RDI algae	1.005205	E10 switchgrass G	0.811045
RDI <i>Camelina</i>	1.013102	E10 willow	1.001565
RDI canola	1.06596	E10 willow G	1.050834
RDI corn	1.011147	E85 corn A	0.318678
RDI <i>Jatropha</i>	1.06345	E85 corn B	0.327946
RDI palm	1.122711	E85 corn C	0.29244
RDI soy	1.098154	E85 corn D	0.262312
RDII algae	1.017503	E85 corn E	0.319889
RDII <i>Camelina</i>	1.024063	E85 corn stover	0.805311
RDII canola	1.061028	E85 corn stover G	0.733083
RDII corn	1.066353	E85 corn/stover A	0.334483
RDII <i>Jatropha</i>	1.028811	E85 corn/stover B	0.311056
RDII palm	1.111896	E85 forage sorghum	0.423953
RDII soy	1.046127	E85 forest residue	1.066466
E10 corn A	0.742906	E85 forest residue G	0.699889
E10 corn B	0.752367	E85 grain sorghum	0.322939
E10 corn C	0.757532	E85 <i>Miscanthus</i>	1.009314
E10 corn D	0.607342	E85 <i>Miscanthus G</i>	0.754069
E10 corn E	0.709096	E85 MSW	1.608481
E10 corn stover	0.895212	E85 poplar	1.010535

E10 corn stover G	1.053193	E85 poplar G	0.754313
E10 corn/stover A	0.733678	E85 sugar cane	0.628125
E10 corn/stover B	0.713644	E85 sweet sorghum A	0.421696
E10 forage sorghum	0.787105	E85 sweet sorghum B	0.310876
E10 forest residue	1.050249	E85 swithgrass	0.634812
E10 forest residue G	1.143864	E85 swithgrass G	0.695675
E10 grain sorghum	0.761435	E85 willow	0.928627
E10 <i>Miscanthus</i>	0.913266	E85 willow G	1.007664

*Corn A: dry mill corn without oil extraction; Corn B: dry mill corn with oil extraction; Corn C: wet milling corn; Corn D: combined dry and wet milling corn; Corn/stover A: integrated corn/stover ethanol (associated with corn); Corn/stover B: integrated corn/stover ethanol (associated with stover); Corn E: generic dry milling corn with oil extraction; Sweet sorghum A: conventional; Sweet sorghum B: integrated.

**Table S30. (Super)efficiency scores obtained by the different biofuels considering the uncertainty in 100 scenarios per DMU .
Part 1**

Fuel	Nominal data	Average	Median	Std. Deviation	Fuel	Nominal data	Average	Median	Std. Deviation
BD20 algae	0.7500	0.7449	0.7502	0.0102	E10 forest residue	1.0502	1.0415	1.0494	0.0122
BD20 <i>Camelina</i>	1.0022	1.0022	1.0022	2.4425E-15	E10 forest residue G	0.6999	0.6880	0.6999	0.0174
BD20 canola	1.0065	1.0065	1.0065	2.4425E-15	E10 grain sorghum	0.7614	0.7491	0.7607	0.0232
BD20 corn	0.8245	0.8245	0.8245	4.3580E-05	E10 <i>Miscanthus</i>	0.9133	0.9034	0.9240	0.0479
BD20 <i>Jatropha</i>	1.0156	1.0154	1.0155	0.0002	E10 <i>Miscanthus</i> G	0.7541	0.7406	0.7541	0.0246
BD20 palm	1.0402	1.0402	1.0402	6.1116E-05	E10 MSW	1.0000	1.0227	1.0198	0.0438
BD20 tallow	1.1633	1.1509	1.1623	0.0180	E10 poplar	1.0000	0.9847	1.0000	0.0358
BD20 soy	1.0450	1.0450	1.0450	1.5545E-15	E10 poplar G	0.7543	0.7406	0.7532	0.0232
E10 corn A	0.7429	0.7240	0.7438	0.0312	E10 sugar cane	1.0016	0.9814	1.0015	0.0596
E10 corn B	0.7524	0.7337	0.7533	0.0315	E10 sweet sorghum A	1.1301	1.1197	1.1299	0.0160
E10 corn C	0.7575	0.7402	0.7587	0.0310	E10 sweet sorghum B	0.6723	0.6610	0.6710	0.0179
E10 corn D	0.6073	0.5973	0.6065	0.0160	E10 switchgrass	0.8202	0.8312	0.8436	0.0317
E10 corn E	0.7091	0.6912	0.7102	0.0281	E10 switchgrass G	0.6957	0.6830	0.6957	0.0182
E10 corn stover	0.8952	0.8987	0.9217	0.0575	E10 willow	1.0016	0.9953	1.0018	0.0263
E10 corn stover G	0.7331	0.7218	0.7341	0.0273	E10 willow G	1.0077	1.0356	1.0308	0.0305
E10 corn/stover A	0.7337	0.7119	0.7358	0.0334	E85 corn A	0.3187	0.3187	0.3187	6.6613E-16
E10 corn/stover B	0.7136	0.6980	0.7142	0.0250	E85 corn B	0.3279	0.3279	0.3279	3.8869E-16
E10 forage sorghum	0.7871	0.7718	0.7887	0.0290	E85 corn C	0.2924	0.2924	0.2924	6.1062E-16

Table S31. (Super)efficiency scores obtained by the different biofuels considering the uncertainty in 100 scenarios per DMU . Part

2

Fuel	Nominal data	Average	Median	Std. Deviation	Fuel	Nominal data	Average	Median	Std. Deviation
E85 corn D	0.2623	0.2623	0.2623	4.4408E-16	E85 switchgrass	0.6348	0.6344	0.6360	0.0330
E85 corn E	0.3199	0.3199	0.3199	5.5511E-17	E85 switchgrass G	0.8110	0.8106	0.8110	0.0040
E85 corn stover	0.8053	0.8204	0.7913	0.0887	E85 willow	0.9286	0.9276	0.9286	0.0028
E85 corn stover G	1.0532	1.0547	1.0532	0.0025	E85 willow G	1.0508	1.0508	1.0507	0.0055
E85 corn/stover A	0.3345	0.3345	0.3345	4.996E-16	RDI algae	1.0052	1.0052	1.0052	4.4409E-16
E85 corn/stover B	0.3111	0.3111	0.3111	4.4408E-16	RDI <i>Camelina</i>	1.0131	1.0131	1.0131	2.4425E-15
E85 forage sorghum	0.4240	0.4240	0.4240	2.2211E-16	RDI canola	1.0660	1.0660	1.0660	6.6613E-16
E85 forest residue	1.0665	1.0665	1.0665	1.5543E-15	RDI corn	1.0111	1.0111	1.0111	4.2018E-05
E85 forest residue G	1.1439	1.1445	1.1445	0.0138	RDI <i>Jatropha</i>	1.0635	1.0635	1.0635	1.9984E-15
E85 grain sorghum	0.3229	0.3229	0.3229	7.7717E-16	RDI palm	1.1227	1.1227	1.1227	1.3323E-15
E85 <i>Miscanthus</i>	1.0093	0.9472	1.0098	0.1230	RDI soy	1.0982	1.0982	1.0982	2.2206E-15
E85 <i>Miscanthus</i> G	1.0036	1.0036	1.0036	4.4409E-16	RDII algae	1.0175	1.0175	1.0175	1.3322E-15
E85 MSW	1.6085	1.5884	1.6079	0.0315	RDII <i>Camelina</i>	1.0241	1.0241	1.0241	1.5543E-15
E85 poplar	1.0105	1.0103	1.0104	0.0011	RDII canola	1.0610	1.0610	1.0610	2.6645E-15
E85 poplar G	0.7701	0.7701	0.7701	0.0014	RDII corn	1.0664	1.0664	1.0664	2.2204E-15
E85 sugar cane	0.6281	0.6206	0.6250	0.0134	RDII <i>Jatropha</i>	1.0288	1.0288	1.0288	1.1102E-15
E85 sweet sorghum A	0.4217	0.4217	0.4217	3.8862E-16	RDII palm	1.1119	1.1119	1.1119	1.3323E-15
E85 sweet sorghum B	0.3109	0.3109	0.3109	3.3307E-16	RDII soy	1.0461	1.0461	1.0461	1.3323E-15

**Table S32. Improvement percentage required for inefficient fuels to become efficient considering the SBM non-oriented model.
 Description of the corn and sorghum process in the Table footnote*.**

Fuel	Inputs			Undesirable outputs								Output
	Cost	LO	Water	GWP	FWEC	FWEU	HT	PMFP	POFP	TA	TE	Distance
BD20 algae	0%	34%	72%	15%	0%	5%	7%	18%	18%	14%	9%	0%
BD20 corn	16%	35%	0%	8%	0%	61%	44%	18%	18%	0%	74%	0%
E10 corn A	0%	45%	95%	12%	11%	28%	18%	5%	6%	0%	63%	0%
E10 corn B	0%	42%	95%	12%	9%	27%	16%	5%	5%	0%	61%	0%
E10 corn C	0%	40%	95%	12%	8%	28%	17%	3%	4%	0%	60%	0%
E10 corn D	0%	95%	98%	1%	27%	28%	26%	8%	8%	5%	94%	6%
E10 corn/stover A	0%	50%	95%	8%	12%	30%	20%	6%	5%	0%	66%	0%
E10 corn/stover B	5%	73%	97%	6%	17%	17%	17%	4%	6%	0%	73%	0%
E10 corn E	1%	64%	96%	8%	17%	30%	21%	6%	6%	0%	73%	0%
E10 switchgrass	0%	94%	53%	0%	0%	1%	0%	0%	2%	1%	47%	0%
E10 <i>Miscanthus</i>	0%	37%	26%	1%	0%	2%	0%	0%	1%	1%	27%	0%
E10 corn stover	0%	57%	6%	1%	0%	3%	0%	1%	4%	2%	41%	0%
E10 grain sorghum	0%	87%	26%	4%	22%	24%	21%	4%	4%	0%	70%	0%
E10 sweet sorghum B	8%	83%	99%	0%	13%	21%	17%	6%	9%	3%	88%	2%
E10 forage sorghum	8%	67%	67%	0%	4%	7%	6%	1%	5%	0%	70%	0%
E85 corn A	0%	99%	98%	55%	48%	70%	69%	49%	54%	33%	98%	22%
E85 corn B	0%	99%	98%	52%	46%	69%	67%	46%	52%	30%	98%	23%
E85 corn C	0%	99%	98%	52%	39%	66%	65%	43%	49%	36%	98%	41%
E85 corn D	0%	99%	98%	42%	41%	58%	60%	35%	45%	25%	99%	72%

E85 corn/stover A	0%	99%	98%	53%	42%	70%	68%	47%	50%	28%	98%	22%
E85 corn/stover B	9%	99%	98%	18%	22%	35%	47%	15%	29%	0%	97%	84%
E85 corn E	0%	99%	98%	52%	45%	68%	66%	45%	50%	26%	98%	28%
E85 willow	0%	0%	5%	8%	0%	12%	12%	11%	11%	16%	3%	0%
E85 switchgrass	0%	0%	53%	29%	37%	50%	44%	31%	32%	50%	76%	0%
E85 corn stover	0%	0%	8%	16%	0%	27%	20%	17%	28%	29%	70%	0%
E85 grain sorghum	0%	99%	99%	54%	54%	72%	73%	53%	60%	42%	99%	11%
E85 sweet sorghum A	41%	99%	99%	2%	0%	50%	53%	32%	52%	23%	97%	19%
E85 sweet sorghum B	36%	99%	99%	0%	10%	46%	44%	26%	43%	16%	97%	71%
E85 forage sorghum	48%	99%	99%	0%	3%	34%	37%	20%	42%	15%	97%	30%
E85 sugarcane	0%	0%	77%	20%	0%	52%	58%	39%	49%	41%	73%	0%
E10 poplar G	4%	97%	98%	5%	5%	0%	1%	1%	6%	0%	52%	4%
E10 switchgrass G	1%	97%	98%	0%	24%	15%	13%	3%	7%	3%	75%	5%
E10 <i>Miscanthus</i> G	0%	97%	92%	0%	18%	6%	4%	1%	4%	0%	48%	6%
E10 corn stover G	0%	97%	84%	0%	21%	11%	8%	0%	4%	0%	69%	5%
E10 forest residue G	0%	97%	92%	0%	24%	14%	10%	4%	8%	4%	77%	5%
E85 poplar G	0%	6%	2%	13%	25%	36%	39%	20%	18%	33%	61%	0%
E85 switchgrass G	12%	28%	34%	11%	17%	18%	18%	22%	15%	25%	20%	6%

*Corn A: Dry mill corn without oil extraction; Corn B: Dry mill corn with oil extraction; Corn C: Wet milling corn; Corn D: combined dry and wet milling corn; Corn/stover A: integrated corn/stover ethanol (associated with corn); Corn/stover B: integrated corn/stover ethanol (associated with stover); Corn E: Gen dry milling corn with oil extraction; Sweet sorghum A: Conventional; Sweet sorghum B: Integrated.

Note that values for inputs and undesirable outputs in Table S30 represent the required reduction despite being shown as positive numbers. Values for outputs denote the required increments.

Table S33. Median improvement percentage required for inefficient fuels to become efficient considering the SBM non-oriented model biofuels considering the uncertainty in 100 scenarios per DMU.

Fuel	Inputs			Undesirable outputs								Output
	Cost	LO	Water	GWP	FWEC	FWEU	HT	PMFP	POFP	TA	TE	Distance
BD20 algae	15%	35%	0%	8%	0%	61%	44%	18%	18%	0%	74%	0%
BD20 corn	0%	34%	72%	15%	0%	5%	7%	18%	18%	14%	9%	0%
E10 corn A	0%	47%	95%	13%	7%	27%	17%	5%	6%	0%	63%	0%
E10 corn B	0%	44%	94%	13%	5%	26%	15%	5%	6%	0%	62%	0%
E10 corn C	0%	42%	94%	13%	4%	26%	16%	3%	5%	0%	60%	0%
E10 corn D	3%	96%	98%	7%	31%	32%	30%	13%	14%	10%	95%	6%
E10 corn/stover A	1%	64%	95%	9%	11%	29%	21%	6%	7%	1%	73%	0%
E10 corn/stover B	4%	79%	97%	7%	19%	18%	19%	5%	7%	1%	77%	0%
E10 corn E	1%	73%	97%	9%	17%	29%	21%	6%	7%	1%	78%	0%
E10 switchgrass	0%	83%	35%	2%	0%	1%	0%	0%	2%	1%	49%	0%
E10 <i>Miscanthus</i>	0%	34%	8%	2%	0%	0%	0%	0%	1%	1%	37%	0%
E10 corn stover	0%	35%	0%	2%	0%	5%	0%	1%	4%	2%	36%	0%
E10 grain sorghum	0%	87%	27%	6%	18%	23%	20%	4%	5%	0%	70%	0%
E10 sweet sorghum B	8%	84%	99%	2%	15%	22%	18%	8%	11%	4%	88%	0%
E10 forage sorghum	6%	67%	67%	0%	4%	7%	6%	1%	5%	0%	70%	0%
E85 corn A	15%	99%	98%	62%	55%	75%	74%	56%	61%	43%	99%	0%

E85 corn B	15%	99%	98%	59%	54%	74%	72%	54%	59%	41%	99%	0%
E85 corn C	29%	99%	98%	66%	57%	76%	75%	60%	64%	54%	99%	0%
E85 corn D	42%	99%	99%	66%	66%	76%	77%	62%	68%	57%	99%	0%
E85 corn/stover A	18%	99%	98%	61%	52%	75%	73%	56%	59%	40%	98%	0%
E85 corn/stover B	51%	99%	99%	56%	58%	65%	71%	54%	61%	46%	98%	0%
E85 corn E	22%	99%	99%	62%	57%	75%	74%	57%	61%	42%	99%	0%
E85 willow	0%	0%	5%	8%	0%	12%	12%	11%	11%	16%	2%	0%
E85 switchgrass	0%	0%	54%	29%	36%	49%	44%	31%	32%	49%	77%	0%
E85 corn stover	0%	0%	13%	18%	0%	28%	22%	23%	32%	34%	65%	0%
E85 grain sorghum	10%	99%	99%	59%	58%	75%	75%	57%	64%	48%	99%	0%
E85 sweet sorghum A	50%	99%	99%	18%	16%	58%	61%	43%	59%	35%	97%	0%
E85 sweet sorghum B	63%	99%	99%	41%	47%	68%	67%	57%	67%	51%	98%	0%
E85 forage sorghum	60%	99%	99%	23%	25%	49%	52%	38%	55%	35%	98%	0%
E85 sugarcane	0%	0%	77%	20%	0%	53%	59%	40%	49%	41%	72%	0%
E10 poplar G	0%	97%	96%	0%	16%	5%	4%	1%	5%	0%	51%	0%
E10 switchgrass G	1%	97%	98%	0%	24%	15%	13%	3%	7%	3%	75%	0%
E10 <i>Miscanthus</i> G	0%	97%	92%	0%	17%	6%	4%	2%	5%	0%	49%	0%
E10 corn stover G	0%	96%	83%	1%	21%	10%	7%	1%	4%	0%	69%	0%
E10 forest residue G	0%	97%	92%	0%	24%	14%	10%	4%	8%	4%	77%	0%
E85 poplar G	0%	6%	2%	13%	25%	36%	39%	20%	18%	33%	61%	0%
E85 switchgrass G	0%	24%	32%	13%	18%	19%	19%	23%	16%	27%	18%	0%

To evaluate the influence of irrigation by rainfall on the efficiencies, the methodology developed in this study was applied to two new possible scenarios as shown in Figure S1 and compared the nominal case that considers the total crop water requirements (i.e., rainwater and irrigation water) and two scenarios that correspond to the state of Oregon and the state of California, with annual rainfall of 922 mm rainfall and 445 mm rainfall respectively. The annual water requirements and the irrigation water supply required by the crops were obtained with the help of CROPWAT software [146]. In both regions, the same loam soil type was considered, and the typical planting months for each crop in these regions were taken into account.

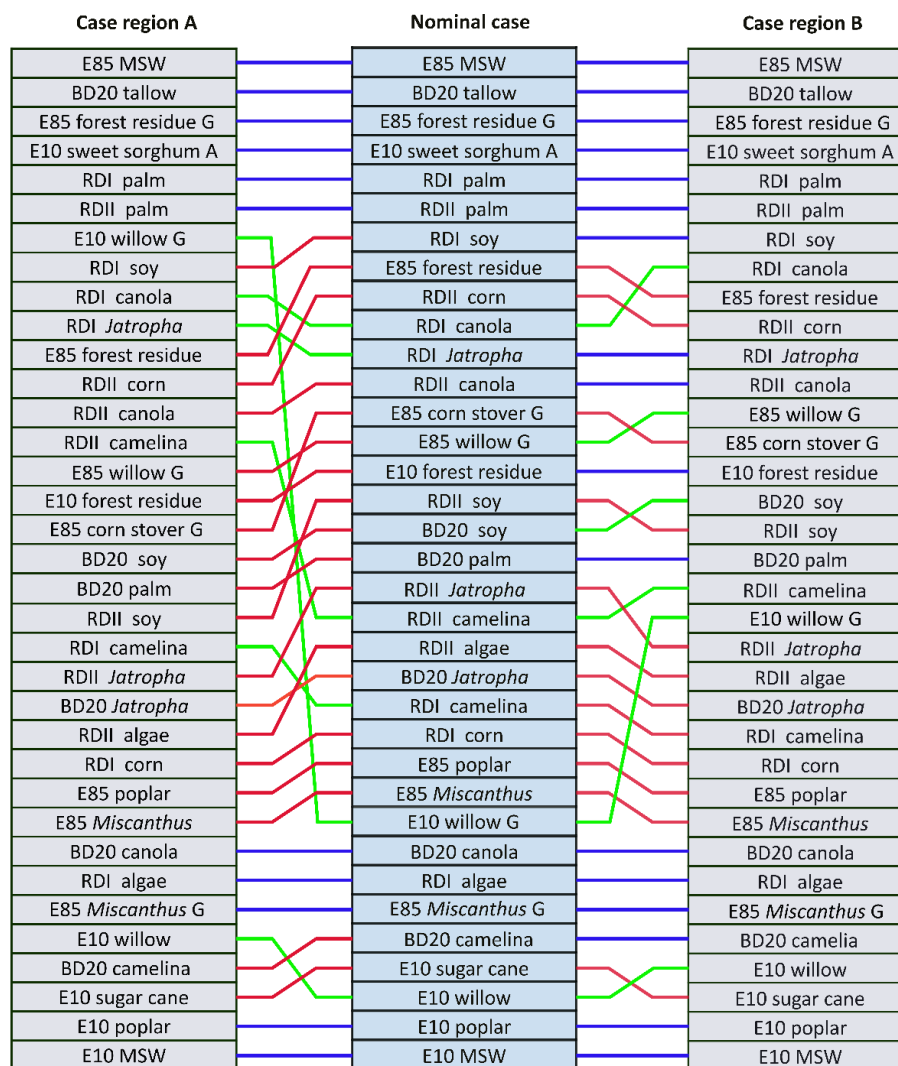


Fig. 9 Variation in efficiency considering the contribution of rain-fed irrigation to the crops

The blue line represents the routes that maintained their position in ranking the efficient DMUs compared to the nominal case. The green line and the red line show an increase and decrease in their position, respectively.

Since the study involves the production of all crops in the same region, they are all influenced by the higher or lower annual rainfall received. However, an improvement is observed in certain crops because their use of rainwater is greater due to their higher water requirements may correspond with rainy periods (e.g., camelina) or because they are annual and can take advantage of the total rainfall throughout the year (e.g., willow).

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Supporting Information

The implications of microalgae biofuel production for the heavy-duty transport sector under planetary boundary perspective

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Number of tables: 32

This document is structured in 3 parts, section 1 provides details about the scenarios, and planetary boundaries (PB) addressed in this paper, section 2 describes the data sources and the LCI developed for the analysis, and section 3 presents additional results not shown in the manuscript for the sake of shortness.

1. Definition of scenarios and planetary boundaries

The scenarios considered in the analysis and the corresponding labels are provided in **Table S1**. In **Table S2**, the abbreviations adopted for the PBs are described, and Table S3 provides the control variables values, along with the proposed boundaries.

Table S34. Correspondence between labels and scenarios reported in this study.

Label	Scenario
BAU	Business-as-usual (global demand for road freight covered by diesel)
M2020	Current global electricity grid mix.[70]
M2040	2040 sustainable development (SD) grid mix. [147]
NGP	Carbon dioxide provided to algae harvesting is captured from the natural gas power plant.[148]
DAC	Carbon dioxide provided to algae harvesting is captured from direct air capture plant.[149]
CCS	Carbon dioxide after cogeneration is captured and stored in a geological reservoir.[150,151]
CCU	Carbon dioxide after cogeneration is captured and sent to algae harvesting. [152]
NoCCU	Remaining Lipid Extracted Algae (LEA) is not used as a source of energy, therefore the use of CO ₂ is not considered and this is considered strictly as an emission
C	Cogeneration of energy occurs through the direct combustion of LEA to supply heat via a steam boiler. Considering an electric efficiency of 21.7% and a heat recovery efficiency of 65% and a biomass moisture of 20%[152]
B	Cogeneration of energy is carried out through anaerobic digestion to produce biogas and its subsequent use in a gas turbine. Considering an electric efficiency of 33% and a heat recovery efficiency of 64%. [152]
ACR	Combustion gases from the cogeneration process are released into the atmosphere without carbon dioxide capture
HDO	Biofuel from hydrodeoxygenation
BD20	Biofuel from transesterification in a blend with fossil fuels 20% vol
HTL	Biofuel from hydrothermal liquefaction

†HTL-Soybean is not considered because for biofuel production by HTL a moisture content of 20% is considered. [153]. Soybean oil is considered a feedstock and currently the soybean residue has an important economic value (i.e., 56% allocation). Therefore, the use of whole grains as feedstock is not considered. [11]

Figure S1 shows in a more detailed representation than Figure 3 of the manuscript, showing the interactions between the different processes in the scenarios under analysis.

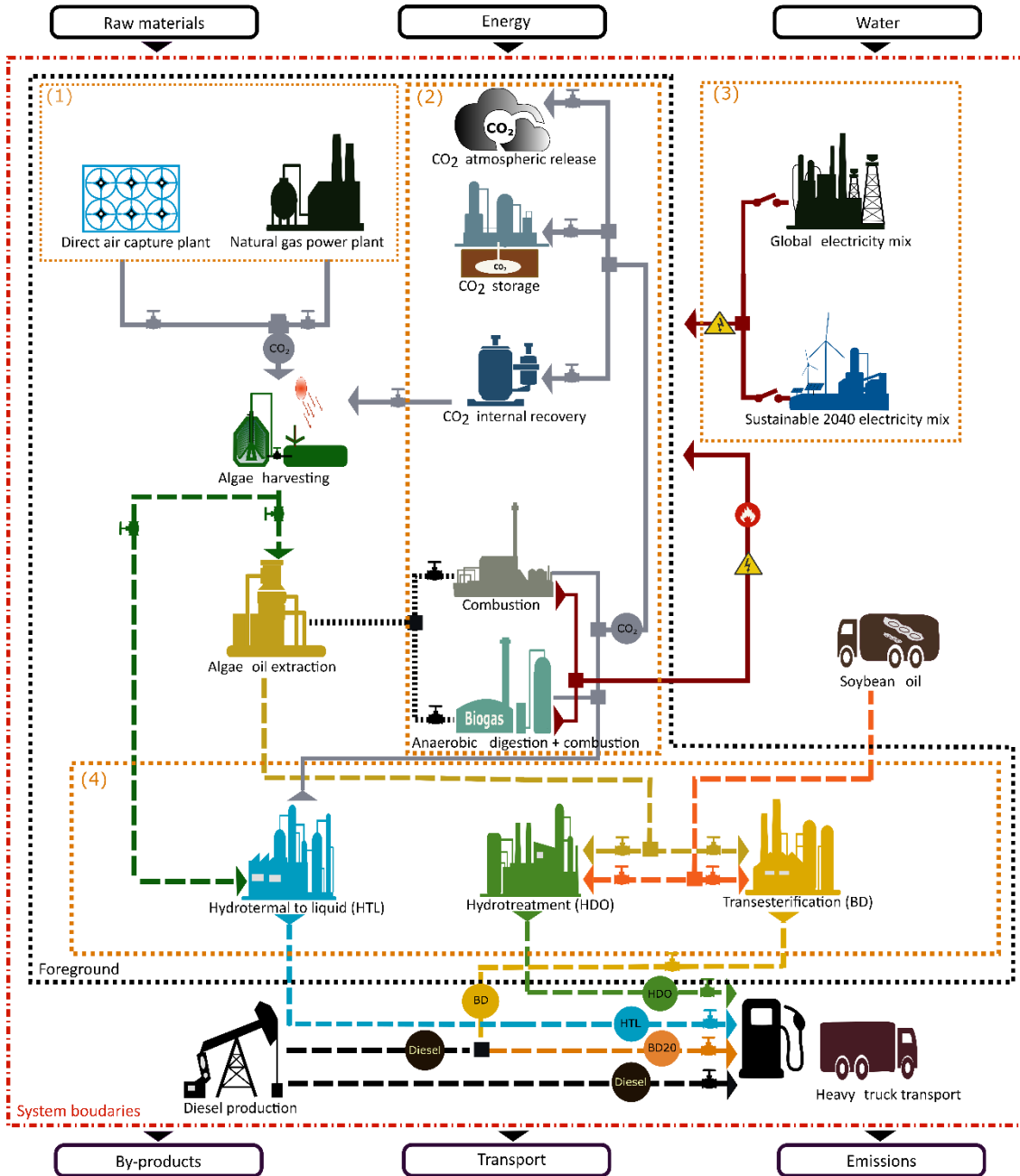


Figure S1. Detailed conceptual framework for biofuels-from microalgae considering 71 scenarios: (1) Carbon feedstock from carbon captured from DAC and power plant; (2) Carbon storage, carbon utilization, and carbon emissions as end life of CO₂ from lipid extracted algae (LEA); (3) Electricity from wind, current global electricity mix, and sustainable mix; (4) Transesterification, hydrotreatment and hydrothermal to liquid as the biofuel conversion process.

Table S35. Abbreviations adopted to denote the control variables associated with the investigated Planetary Boundaries.

Label	Planetary boundary or earth system process	Control variable
aCO ₂	Climate change	CO ₂ atmospheric

EI	Climate change	concentration [ppm]
SOD	Stratospheric ozone depletion	Energy imbalance [$W m^{-2}$] Stratospheric ozone concentration [Dobson units, DU]
OA	Ocean acidification	Carbonate ion concentration [Ω_{arag}]
P	Biogeochemical flow	Phosphorus flow from freshwater systems into the ocean [$Tg P a^{-1}$]
N	Biogeochemical flow	Industrial and intentional biological fixation of N [$Tg N a^{-1}$]
LSC	Land system change	Area of forested land [% of original forest cover]
FWU	Freshwater use	Maximum amount of consumptive blue water use [$km^3 a^{-1}$]
CBI	Terrestrial biosphere integrity	Functional diversity [Biodiversity Intactness Index, BII]

Table S3 Control variables values, along with the proposed boundaries considering the whole economy.

Label	Unit	Planetary boundary	Zone of uncertainty	Natural background level	Safe operating space	Current anthropogenic status
aCO ₂	ppm CO ₂	350	350 - 450	278	72	398.5
EI	$W m^{-2}$	1	1-1.5	0	1	2.3
SOD	DU	275	275 -261	290	15	200
OA	Ω_{arag}	2.75	2.75-2.4	3.44	0.69	2.89
P	$Tg P year^{-1}$	11	11-100	1.1	9.9	22
N	$Tg N year^{-1}$	62	62-82	0	62	150
LSC	%	75	75-54	100	25	62
FWU	$km^3 year^{-1}$	4000	4000-6000	0	4000	2600
CBI	BII loss	10	10-70	0	10	26.8

2. Data sources and the LCI developed

2.1. Data sources

The required data used in the LCA calculations is provided in this section. Most values were taken from the GREET database, [152] which contains harmonized data from 1400 facilities with different annual capacities, as shown in Table S4.

Table S4. Annual capacity of plants considered for the LCI data gathered.

Facility	Annual capacity	Reference
Microalgae cultivation pond	350 ton dry algae	[154]
Microalgae cultivation facility	1750 ton dry algae	[154]
Biofuel plant	42500 ton diesel	[155]
CO ₂ for direct air capture	1.08 ton CO ₂	[149]
CO ₂ capture and storage in a geological reservoir	8.7 ton CO ₂	[151]

Table S5 to Table S21 provide the different life cycle inventories considered for microalgae biofuel production, along with all the dependent sub-processes that were adapted from the original references. The final LCIs were obtained by combining data from the process model in GREET with information from the literature and Ecoinvent v3.7. [70] The latter activities, missing in Ecoinvent and requiring a tailored intermediate inventory analysis based on the literature, are labeled with a "*" detailed in LCI Tables. Furthermore, as seen in tables S5 and S21, several of the inputs to the foreground system were modeled utilizing additional information from the literature and Ecoinvent.

The "Electricity" activity differs depending on the scenario. Table S22 and Table S23 provide a thorough breakdown of the various electricity scenario options. We assume that the electricity consumed in the background system is modeled with the default activities in Ecoinvent v3.7. Hence, we only change the electricity of the foreground system while keeping the energy generation activities in the background system unaltered.

Table S5-S7 provide information about microalgae production with a moisture of 20%. The process considers the use of open pools, where water and fertilizers are recirculated after the filtration and drying process so that the replacement water introduced corresponds to the water lost by evaporation and blowdown to avoid excess mineral and salt build-up, and to regulate the pH of the culture medium. [156] The harvesting and drying process is carried out in two stages using flocculation followed by mechanical filtration using centrifuges [152].

The yield of microalgae is influenced by their geographical location and, as a result, the potential of microalgae to reduce their impacts on seven Earth-system processes can vary across different scenarios. The yield of microalgae varies widely across the world, ranging from 4.43 g/m²/day (1.13 m³ oil/ha/yr) to 38.80 g/m²/day (27 m³/ha/yr), with the geographical location and season affecting growth due to variations in radiation and temperature. [157] However, for the purposes of this study, harmonized values corresponding to 26.37 g/m²/day (3.6 m³/ha/yr) were used, based on studies conducted in the United States region. [154]

The process of CO₂ capture from direct air capture (Table S15) or natural gas power plant (Table S15) provides concentrations higher than 97%. This concentration is adopted since GREET describes a pure CO₂

flow for microalgae growing. Furthermore, in the case of CO₂ from the natural gas power plant to remove compounds such as nitrogen oxides, sulfur oxides and heavy metals that can adversely affect the growth of microalgae.

Table S5. LCI of the foreground system microalgae production based on open ponds excluding the recovery of CO₂ after energy cogeneration based on lipid extracted algae (LEA).

Process: Microalgae production w/o CO ₂ from downstream activities			
Ecoinvent entry	Description	Amount	
Inputs:			
	Electricity cultivation and drying [kWh]	0.614	
Market for electricity medium voltage			
Nutrient supply from urea, ROW	Nitrogen fertilizer [kg]	0.025	
Market for sodium phosphate glo	Phosphorus fertilizer [kg]	0.010	
Aluminium sulfate, powder//[ROW] market for aluminium sulfate, powder	Flocculant [kg]	0.004	
Concrete block//[RoW] market for concrete block	Concrete [kg]	0.023	
Market for reinforcing steel	Steel [kg]	0.001	
Extrusion, plastic film//[GLO] market for extrusion, plastic film	Plastic [kg]	0.005	
Cast iron//[GLO] market for cast iron	Cast iron [kg]	0.003	
Water, unspecified natural origin, RoW	Water lost to blowdown and evaporation [kg]	57.3	
Occupation, unspecified, in ground	Land occupation [m ² a]	0.00013	
*CO ₂ from NG or DAC	CO ₂ input [kg]	2.67	
Outputs:			
	Microalgae biomass 20% moisture[kg]	1	
	CO ₂ emissions [kg]	0.459	

*CO₂ from NG and DAC are the possible activities that supply CO₂ to the process and are detailed on Table S15 and Table S16, respectively. The LCI data for microalgae biomass production are retrieved from the GREET2022 datasheet in section 1.3 of the "Algae" category. [158] More information and details on cultivation can also be found in the document "2017 Algae Harmonization Study". [154]

Table S6. LCI of the foreground system microalgae production based on open ponds with CO₂ recovery after LEA combustion for energy cogeneration.

Process: Microalgae production w CO ₂ from LEA combustion		
Ecoinvent entry	Description	Amount

Inputs:		
	Electricity Cultivation and	
Market for electricity medium voltage	drying [kWh]	0.293
Nutrient supply from urea, ROW	Nitrogen fertilizer [kg]	0.025
Market for sodium phosphate [GLO]	Phosphorus fertilizer [kg]	0.010
Aluminium sulfate, powder//[ROW] market for aluminium sulfate, powder	Flocculant [kg]	0.004
Concrete block//[RoW] market for concrete block	Concrete [kg]	0.023
Market for reinforcing steel	Steel [kg]	0.001
Extrusion, plastic film//[GLO] market for extrusion, plastic film	Plastic [kg]	0.005
Cast iron//[GLO] market for cast iron	Cast iron [kg]	0.003
Water, unspecified natural origin, RoW	Water lost to blowdown and evaporation [kg]	57.3
Occupation, unspecified, in ground	Land occupation [m ² a]	0.00013
*CO ₂ from NG or DAC	CO ₂ input [kg]	1.229

Outputs:		
	Microalgae biomass 20% moisture [kg]	1
	CO ₂ emissions [kg]	0.459

†For CO₂ reuse from LEA combustion, 1.23 kg CO₂/kg algae biomass is supplied, 0.336 kWh of electricity is delivered from the cogeneration plant, and 0.015kWh are required for capture and transport of CO₂ from LEA combustion with a moisture of 20%. The LCI data for microalgae biomass production are retrieved from the GREET2022 datasheet in section 1.3 of the "Algae" category.[158] More information and details on cultivation can also be found in the document "2017 Algae Harmonization Study".[154] The electric and heat recovery efficiencies are 21.7% and 65%, respectively.[152,159]

*CO₂ from NG and DAC are the possible activities that supply CO₂ to the process and are detailed in Table S14 and Table S15, respectively.

Table S7. LCI of the foreground system microalgae production based on open ponds with CO₂ recovery after LEA combustion for energy cogeneration.

Process: Microalgae production w CO ₂ from biogas combustion		
Ecoinvent entry	Description	Amount
Inputs:		
	Electricity cultivation and	
Market for electricity medium voltage	drying [kWh]	0.0368
Nutrient supply from urea, ROW	Nitrogen fertilizer [kg]	0.025
Market for sodium phosphate glo	Phosphorus fertilizer [kg]	0.010

Aluminium sulfate, powder//[ROW] market for aluminium sulfate, powder	Flocculant [kg]	0.004
Concrete block//[RoW] market for concrete block	Concrete [kg]	0.023
Market for reinforcing steel	Steel [kg]	0.001
Extrusion, plastic film//[GLO] market for extrusion, plastic film	Plastic [kg]	0.005
Cast iron//[GLO] market for cast iron	Cast iron [kg]	0.003
Water, unspecified natural origin, RoW	Water lost to blowdown and evaporation [kg]	57.3
Occupation, unspecified, in ground	Land occupation [m ² a]	0.00013
*CO ₂ from NG or DAC	CO ₂ input [kg]	1.77

Outputs:

Microalgae biomass 20% moisture [kg]	1
CO ₂ emissions [kg]	0.459

For CO₂ reuse from biogas combustion, 0.69 kg of CO₂ are supplied for each kg of algae introduced to the microalgae production process, 0.592 kWh of electricity are delivered from the cogeneration plant, and 0.008 kWh are required for capture and transport of CO₂ from biogas combustion. The electric and heat recovery efficiencies are 33.1% and 64%, respectively for combined heat and power through a gas turbine. [152,159,160].

*CO₂ from NG and DAC are the possible activities that supply CO₂ to the process and are detailed on table Table S15 and Table S16, respectively.

Table S8. LCI of the microalgae oil production based on wet extraction without cogeneration.

Process: Microalgae oil extraction			
Ecoinvent entry	Description	Amount	
Inputs:			
*Algae biomass 20% moisture	Algae biomass 20% moisture [kg]	4.678	
Heat, central or small-scale, other than natural gas {GLO} market group for APOS, U	Thermal energy for extraction [MJ]	11.124	
Electricity, medium voltage {GLO} market group for APOS, U	Electricity for extraction [kWh]	0.927	
Hexane {GLO} market for APOS, U	Hexane [kg]	5.50E-05	
Outputs:			
	Lipid-extracted biomass to recovery step [kg]	3.98	

Hexane emissions [kg]	5.50E-05
Microalgae oil [kg]	1

The data for microalgae extraction are retrieved from the GREET2022 datasheet in section 2.3 from oil extraction column, of the "Algae" category. [158]

Table S9. LCI of the microalgae oil production based on wet extraction.

Process: Microalgae oil extraction			
Ecoinvent entry	Description	Amount	
Inputs:			
*Algae biomass 20% moisture	Algae biomass 20% moisture [kg]	4.678	
Heat, central or small-scale, other than natural gas {GLO} market group for APOS, U	Thermal energy for extraction [MJ]	0	
Electricity, medium voltage {GLO} market group for APOS, U	Electricity for extraction [kWh]	0.927	
Hexane {GLO} market for APOS, U	Hexane [kg]	5.50E-05	
Outputs:			
	Lipid-extracted biomass to recovery step [kg]	3.98	
	Hexane emissions [kg]	5.50E-05	
	Microalgae oil [kg]	1	

†The heat demand is considered 0 for the scenarios with cogeneration of energy by direct combustion of LEA or cogeneration from biogas. In the first case, 23.12 MW/kg oil, and in the second case, 17.75 MW/ kg oil of thermal energy are generated [152].

Table S10. LCI of the energy cogeneration from lipid extracted algae combustion.

Process: Energy cogeneration from LEA combustion			
Ecoinvent entry	Description	Amount	
Inputs:			
Lipid-extracted biomass to recovery step	Lipid-extracted biomass [kg]	3.98	
Outputs:			
	Thermal energy recovery [MJ]	23.12	
	Electricity recovery [kWh]	2.73	
	CO ₂ recovered [kg]	5.76	
	CO ₂ emissions [kg]	1.265	

LCI data for LEA combustion are retrieved from the GREET2022 datasheet in section 2.1 of the "Algae" category where 100% LEA to combustion is selected. [158]

Table S11. LCI of the energy cogeneration from biogas combustion.

Process: Energy cogeneration from biogas			
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Ecoinvent entry	Description	Amount
Inputs:		
*Lipid-extracted biomass	Lipid-extracted biomass [kg]	3.98
Outputs:		
	Thermal energy recovery [MJ]	17.75
	Electricity recovery [kWh]	3.81
	CO ₂ recovered [kg]	2.33
	Anaerobic digestate residue [kg]	1.99
	Methane emissions [g]	16.96

LCI data for LEA combustion are retrieved from the GREET2022 datasheet in section 2.1 of the "Algae" category where 100% LEA to biogas combustion is selected. [158]

Table S12. LCI of the HDO100 from microalgae production.

Process: HDO100 production from microalgae		
Ecoinvent entry	Description	Amount
Inputs:		
Electricity, medium voltage {GLO} market group for APOS, U	Electricity [kWh]	0.086
*Microalgae oil	Microalgae oil [kg]	0.962
Tap water {Europe without Switzerland} tap water production, conventional treatment APOS, U	Water [kg]	0.0077
Hydrogen, liquid {RER} market for APOS, U	Hydrogen [kg]	0.031
Outputs:		
	HDO [kg]	1
	Fuel gas [kg]	0.253
	Heavy oil [kg]	0.175

LCI data for HDO production are retrieved from the GREET2022 datasheet in section 2.2 of the "Algae" category where "Renewable Diesel I" column is selected. [158]

†The economic allocation considered was 89% for HDO, 6% for fuel gas, and 5% for heavy oil.

Table S13. LCI of the BD100 from microalgae production.

Process: BD100 production from microalgae		
Ecoinvent entry	Description	Amount
Inputs:		
Heat, central or small-scale, other than natural gas {GLO} market group for APOS, U	Thermal energy [MJ]	1.07

Electricity, medium voltage {GLO} market group for APOS, U	Electricity [kWh]	0.0335
Tap water {Europe without Switzerland} tap water production, conventional treatment APOS, U	Water [kg]	0.0076
Methanol {GLO} market for APOS, U	Methanol [kg]	0.109
Sodium methoxide {GLO} market for APOS, U	Sodium methoxide [kg]	0.0040
Neutralising agent, sodium hydroxide-equivalent {GLO} market for APOS, U	Sodium hydroxide [kg]	0.0011
Phosphoric acid, industrial grade, without water, in 85% solution state {GLO} market for APOS, U	Phosphoric acid [kg]	0.0004
Hydrochloric acid, without water, in 30% solution state {RoW} market for APOS, U	Hydrochloric acid [kg]	0.0017
Sulfuric acid {RoW} market for sulfuric acid APOS, U	Sulfuric acid [kg]	0.0011
*Microalgae oil	Microalgae oil [kg]	0.958
Outputs:		
	Biofuel [kg]	1
	Glycerin [kg]	0.097

LCI data for biodiesel production are retrieved from the GREET2022 datasheet in section 2.2 of the "Algae" category where "Biodiesel" column is selected. [158]

†Economic allocation of 95.7% for biofuel and 4.3% for glycerin were considered.

Table S14. LCI of the HTL from microalgae production.

Process: HTL100 production from microalgae			
	Ecoinvent entry	Description	Amount
Inputs:			
	Heat, district or industrial, natural gas {GLO} market group for APOS, U	Heat [MJ]	8.516
	Electricity, medium voltage {GLO} market group for APOS, U	Electricity [kWh]	0.190
	Tap water {Europe without Switzerland} tap water production, conventional treatment APOS, U	Water [kg]	0.647
	Wood chips, dry, measured as dry mass {RoW} market for APOS, U	Wood [kg]	0.642
	*Microalgae biomass	Microalgae biomass	1.613

		[kg]	
Sulfuric acid {RER} market for sulfuric acid			
APOS, U	Sulfuric acid [kg]	0.076	
*HTL catalyst	Catalyst [kg]	0.000122	
Outputs:			
	HTL [kg]	0.673	
	Gasoline [kg]	0.327	
	CO ₂ emissions [kg]	2.495	

LCI data for HTL production are retrieved from the GREET2022 datasheet in section 2.2 of the "Algae" category where "Hydrothermal liquefaction" column is selected. [158] More information and details on cultivation can also be found in the document "2017 Algae Harmonization Study". [154]

*LCI of catalyst are detailed in Table S21.

†Economic allocation of 67.3% for HTL and 32.7% for gasoline were considered.

Table S15. LCI of the CO₂ from direct air capture plant. [149]

Process:		CO ₂ from direct air capture plant	
Ecoinvent entry	Description	Amount	
Inputs:			
Heat, district or industrial, natural gas {GLO} market group for	Natural gas [MJ]	5.25	
Electricity, high voltage {GLO} market group for	Electricity [kWh]	0.234	
Tap water {GLO} market group for	Water [kg]	3.11	
Calcium carbonate, precipitated {RoW} market for calcium carbonate, precipitated	Calcium carbonate [kg]	0.0199	
Outputs:			
	CO ₂ captured [kg]	1	

LCI data for CO₂ capture from direct air capture plant was modeled in Ecoinvent based on the data provided by Keith et al. [149]

Table S16. LCI of the CO₂ capture from natural gas power plant.

Process:		CO ₂ capture from natural gas power plant	
Ecoinvent entry	Description	Amount	
Inputs:			
Natural gas, from high pressure network (1- 5 bar), at service station {GLO} market for	Natural gas [kg]	4.78E-01	
Water, decarbonized, at user {RER} water production and supply, decarbonized	Water [kg]	3.30E-01	
Water, unspecified natural origin {GLO}	Water [kg]	1.45E-03	
*Catalyst	Catalyst [kg]	2.97E-06	
Monoethanolamine {GLO} market for	Solvent MEA [kg]	8.77E-03	

Outputs:	
Carbon dioxide [kg]	1
Electricity [kWh]	3.28
CO ₂ emissions [kg]	3.29E-01
Monoethanolamine emissions [kg]	3.52E-03
Nitrogen oxides [kg]	1.58E-03

†LCI of catalyst are detailed in Table S20. LCI data for CO₂ captured from a natural gas power plant was modeled in Ecoinvent based on the data provided by Ioannou et al. [161] The electricity shown is equal to the required demand minus the electricity for compression because compression at 30 bar is not required. [161,162] An economical allocation factor of 81% for electricity and 19% for CO₂ was considered. [148,163]

Table S17. LCI of a pipeline for supercritical CO₂ transport. [151]

Process:	Pipeline supercritical CO ₂		
	Ecoinvent entry	Description	Amount
Inputs:			
	Occupation, construction site	Land occupation [m ² a]	3330
	Transformation from forest	Land transformation [m ²]	2000
	Transformation, to heterogeneous, agricultural	Land transformation [m ²]	2000
	Water, unspecified natural origin/m ³	Water [m ³]	187
	Sand {GLO} market for	Sand [kg]	4.40E+06
	Diesel, burned in building machine {GLO}	Diesel [MJ]	3.31E+06
	Steel, low-alloyed {GLO} market for	Steel [kg]	2.70E+05
	Drawing of pipe, steel {RER} processing	Drawing of pipelines [kg]	2.70E+05
	Stone wool, packed {CH} stone wool production, packed	Rock wool [kg]	5119
	Transport, helicopter {GLO} market for APOS, U	Transport, helicopter [hr]	26
	Transport, helicopter, LTO cycle {GLO} market for APOS, U	Transport helicopter [p]	10.4
	Transport, freight, lorry 16-32 metric ton, euro6 {RER} market for transport, freight, lorry 16-32 metric ton, EURO6	Transport lorry [tkm]	3.15E+05
	Transport, freight train {RER} market group for transport, freight train APOS, U	Transport rail [tkm]	5.51E+04
	Disposal, inert waste 5% water to inert material landfill	Disposal of inert waste [kg]	4.40E+06
	Inert waste, for final disposal {CH} treatment of inert waste, inert material landfill APOS, U	Disposal, steel [kg]	1.35E+05
	Waste mineral wool, for final disposal {CH} treatment of waste mineral wool, inert material landfill APOS, U	Disposal, mineral wool [kg]	5.12E+03
Outputs:		Pipeline supercritical [km]	1

LCI data for the pipeline construction to transport CO₂ for geological storage was modeled in Ecoinvent based on the data provided by Wildbolz et al. [151].

Table S18. LCI for CO₂ transport through pipeline [151].

Process:	Storage CO ₂ aquifer 200 km pipeline		
	Ecoinvent entry	Description	Amount

Inputs:			
	*Well double aquifer	well double aquifer [p]	2.54E-11
	*CO ₂ transport	CO ₂ transport [km]	6.34E-9
Outputs:			
		CO ₂ stored [kg]	1

LCI data for the storage of CO₂ transported in a well double aquifer was modeled in Ecoinvent based on the data provided by Wildbolz et al. [151]

Table S19. LCI for CO₂ capture in a geological reservoir [151]

Process:	CO ₂ capture in a well double aquifer		
	Ecoinvent entry	Description	Amount
Inputs:			
	Occupation, industrial area	Industrial area [m ² a]	900
	Occupation, industrial area, vegetation	Vegetation occupation [m ² a]	8100
	Transformation, from pasture and meadow	Land transformation [m ²]	600
	Transformation, to industrial area	Land transformation [m ²]	60
	Transformation, to industrial area, vegetation	Land transformation [m ²]	540
	Deep well, drilled, for geothermal power {GLO} market for APOS, U	Drilling [m]	3.60E+03
	Cement, unspecified {CH} market for cement, unspecified APOS, U	Cement [kg]	1.26E+05
	Gravel, crushed {CH} production APOS, U	Gravel [kg]	1.32E+06
	Transport, freight, lorry 16-32 metric ton, euro6 {RoW} market for transport, freight, lorry 16-32 metric ton, EURO6 APOS, U	Transport lorry [tkm]	2.89E+04
	Transport, freight train {GLO} market group for APOS, U	Transport train [tkm]	1.26E+04
Outputs:			
		Well double aquifer [p]	1

LCI data for the use of a geological reservoir as CO₂ storage site was modeled in Ecoinvent based on the data provided by Wildbolz et al. [151].

Table S20. LCI of CO₂ capture and compression plant.

Process:	CO ₂ capture and compression for storage		
	Ecoinvent entry	Description	Amount
Inputs:			
	Monoethanolamine {GLO} market for APOS, U	MEA [kg]	3.30E-04
	Water, decarbonised, at user {GLO} market for APOS, U	Water [kg]	6.26E-01
	Electricity, medium voltage {GLO} market group for APOS, U	Pumping [kWh]	3.46E-02
	Electricity, medium voltage {GLO} market group for APOS, U	Compression [kg]	9.24E-02
	Heat, district or industrial, natural gas {GLO} market group for APOS, U	Heating [MJ]	7.52E+00
	Storage CO ₂ aquifer 200 km pipeline	Storage [kg]	1
Outputs:			
		CO ₂ capture [kg]	1.00E+00
		CO ₂ emissions [kg]	3.23E-03
		H ₂ O emissions [kg]	1.21E+00
		O ₂ emissions [kg]	8.27E-01
		N ₂ emissions [kg]	4.44E+00

MEA emissions [kg]	1.17E-04
LCI data for CO ₂ capture and compression plant was modeled in Ecoinvent based on the data provided by Bello et al. [150]	

*For electricity inputs GLO electricity is going to be changed to electricity M2040.

Table S21. LCI of catalyst

Process:	Catalyst		
	Ecoinvent entry	Description	Amount
Inputs:			
	Cobalt {GLO} market for APOS, U	Cobalt oxide [kg]	0.045
	Molybdenum trioxide {GLO} market for APOS, U	Molybdenum trioxide [kg]	0.145
	Aluminium oxide {GLO} market for APOS, U	Alumina [kg]	0.81
Outputs:			
		Catalyst [kg]	1

LCI of catalyst was modeled in Ecoinvent based on the data provided by Albrecht et. Al, [164] which studies the catalytic hydrotreating and hydrothermal liquefaction of microalgae.

Table S22. LCI of sustainable 2040 electricity mix. [147]

Process:	M2040 electricity mix		
	Ecoinvent entry	Description	Amount
Inputs:			
	Electricity, high voltage {RoW} electricity production, hard coal APOS, U	Energy from coal [kWh]	0.0092
	Electricity, high voltage {RoW} electricity production, oil APOS, U	Energy from oil [kWh]	0.0018
	Electricity, high voltage {RoW} electricity production, natural gas, conventional power plant APOS, U	Energy from natural gas [kWh]	0.103
	Electricity, high voltage {RoW} electricity production, nuclear, pressure water reactor APOS, U	Energy from nuclear source [kWh]	0.1598
	Electricity, high voltage {RoW} electricity production, hydro, run-of-river APOS, U	Energy from hydropower [kWh]	0.1583
	Electricity, high voltage {RoW} heat and power co-generation, wood chips, 6667 kW, state-of-the-art 2014 APOS, U	Energy from biomass [kWh]	0.0885
	Electricity, high voltage {RoW} electricity production, wind, 1-3MW turbine, onshore APOS, U	Energy from wind [kWh]	0.3572
	Electricity, high voltage {RoW} electricity production, deep geothermal APOS, U	Energy from geothermal [kWh]	0.0099
	Electricity, low voltage {RoW} electricity production, photovoltaic, 570kWp open ground installation, multi-Si APOS, U	Energy from photovoltaic [kWh]	0.1038
	Electricity, high voltage {RoW} electricity production, solar thermal parabolic trough, 50 MW APOS, U	Energy from solar thermal [kWh]	0.0085
Outputs:			
		Electricity mix M2040 [kWh]	1

LCI of global 2040 electricity mix was modeled in Ecoinvent according to the World Energy Outlook 2019 [147] projection for electricity mix in 2040.

Table S23. LCI of Global electricity mix. The grid mix considers the global share of different countries, according to the Ecoinvent database [70].

Process:		M2040 electricity mix	
	Ecoinvent entry	Description	Amount
Inputs:			
	Electricity, medium voltage {AU} market for APOS, U	Energy from coal [kWh]	0.010
	Electricity, medium voltage {NZ} market for electricity, medium voltage APOS, U	Energy from oil [kWh]	0.002
	Electricity, medium voltage {RAF} market group for APOS, U	Energy from natural gas [kWh]	0.032
	Electricity, medium voltage {RAS} market group for APOS, U	Energy from nuclear source [kWh]	0.461
	Electricity, medium voltage {RER} market group for APOS, U	Energy from hydropower [kWh]	0.173
	Electricity, medium voltage {RLA} market group for APOS, U	Energy from biomass [kWh]	0.062
	Electricity, medium voltage {RNA} market group for APOS, U	Energy from wind [kWh]	0.216
	Electricity, medium voltage {RoW} market for APOS, U	Energy from geothermal [kWh]	0.00008
	Electricity, medium voltage {RU} market for APOS, U	Energy from photovoltaic [kWh]	0.044
Outputs:			
		Electricity mix M2040 [kWh]	1

For combustion, the fuel consumption of 0.023 kg/t km was considered. [152,165] This value is within the range reported with fuel consumption at full load capacity for short-haul trucks between 0.018 and 0.024 kg diesel/t km. [166] In this case, our study, for the sake on simplicity will consider the same fuel consumption per t km, although this value has certain variations according to the type of fuel used.

Table S24. LCI of HDO100 combustion for freight road transport.

Process:		HDO100 combustion for freight road transport	
	Ecoinvent entry	Description	Amount
Inputs:			
	*HDO100 fuel	HDO100 fuel [kg]	0.023
Outputs:			
		Freight road transport [t km]	1
Emissions to air		VOC, volatile organic compounds as C [kg]	2.17E-06
		Carbon monoxide, fossil [kg]	2.25E-05
		Nitrogen oxides [kg]	5.51E-05
		Particulates, < 10 um [kg]	1.24E-06
		Particulates, < 2.5 um [kg]	1.14E-06
		Sulfur oxides [kg]	0.00E+00
		Methane [kg]	2.34E-06
		Carbon dioxide, fossil [kg]	7.32E-02
		Dinitrogen monoxide [kg]	9.55E-08
		Carbon black [kg]	1.02E-07

LCI of HDO100 combustion was modeled in Ecoinvent based on the data provided by GREET® 2021 .Net software, emissions correspond to the combustion of a heavy-duty truck denominated “HD Truck: short haul” powered by HDO100 or renewable diesel.[167]

Table S25. LCI of diesel combustion for freight road transport.

Process:		DIESEL	
	Ecoinvent entry	Description	Amount
Inputs:			
	*Diesel fuel	Diesel fuel [kg]	0.023
Outputs:			
	Emissions to air	Freight road transport [t km]	1
		VOC, volatile organic compounds as C [kg]	2.17E-06
		Carbon monoxide, fossil [kg]	2.25E-05
		Nitrogen oxides [kg]	5.51E-05
		Particulates, < 10 um [kg]	1.24E-06
		Particulates, < 2.5 um [kg]	1.14E-06
		Sulfur oxides [kg]	5.21E-07
		Methane [kg]	2.34E-06
		Carbon dioxide, fossil [kg]	7.55E-02
		Dinitrogen monoxide [kg]	9.55E-08
		Carbon black [kg]	1.02E-07

LCI of diesel combustion was modeled in Ecoinvent based on the data provided by GREET® 2021 .Net software, emissions correspond to the combustion of a heavy-duty truck denominated “HD Truck: short haul” powered by low sulfur diesel.[167]

Table S26. LCI of biodiesel combustion for freight road transport.

Process:		BD20	
	Ecoinvent entry	Description	Amount
Inputs:			
	*BD20 fuel	BD20 fuel [kg]	0.023
Outputs:			
	Emissions to air	Freight road transport [tkm]	1
		VOC, volatile organic compounds as C [kg]	2.17E-06
		Carbon monoxide, fossil [kg]	2.25E-05
		Nitrogen oxides [kg]	5.51E-05
		Particulates, < 10 um [kg]	1.24E-06
		Particulates, < 2.5 um [kg]	1.14E-06
		Sulfur oxides [kg]	4.17E-07
		Methane [kg]	2.34E-06
		Carbon dioxide, fossil [kg]	7.51E-02
		Dinitrogen monoxide [kg]	9.55E-08
		Carbon black [kg]	1.02E-07

LCI of BD20 combustion was modeled in Ecoinvent based on the data provided by GREET® 2021 .Net software, emissions correspond to the combustion of a heavy-duty truck denominated “HD Truck: short haul” powered by biodiesel 20% vol.[167]

3. Additional data

3.1. Overall level of transgression

The results shown in Figure 3 from the main manuscript are analyzed in greater detail for the different scenarios studied (Table S27 to Table S31). The planetary boundaries main contributions activities are detailed from Figure S1 to Figure S9, the resulting datasets generated during the current work are publicly available online at Cabrera-Jimenez R. et al.[168]

Table S27. Share of the global SOS, carbon footprint, human health impacts, and PB footprint for biodiesel production occupied by the global heavy-duty transport sector considering the current global electricity mix, 2040 sustainable electricity mix, and wind electricity.

Fuel	Scenario	aCO ₂	EI	SOD	OA	P	N	LSC	FWU	CBI	CF	HH	PBF
		ppm	Wm ⁻²	DU	Ω _{arag}	Tg P	Tg N	% LSC	km ³	% BII loss	kg CO ₂ eq	DALY	
BD20	BLUE ^{M2020} _{C-CCS}	156%	148%	0%	9.99%	0.02%	1.06%	0.00%	1.26%	9.05%	4.3	752	5.06
BD20	BLUE ^{M2020} _{C-CCU}	141%	134%	0%	9.03%	0.02%	0.61%	0.00%	1.22%	8.09%	3.9	655	5.02
BD20	GREY ^{M2020} _{NoCCU}	173%	164%	0%	11.04%	0.02%	1.02%	0.00%	1.26%	10.02%	4.8	833	5.10
BD20	GREEN ^{M2020} _{C-CCS}	110%	106%	0%	7.07%	0.02%	0.46%	0.00%	1.43%	6.62%	3.1	668	4.95
BD20	GREEN ^{M2020} _{C-CCS}	118%	112%	0%	7.54%	0.02%	0.31%	0.00%	1.31%	6.85%	3.3	612	4.96
BD20	YELLOW ^{M2020} _{NoCCU}	127%	121%	0%	8.11%	0.02%	0.42%	0.00%	1.43%	7.58%	3.6	750	4.99
BD20	BLUE ^{M2040} _{C-CCS}	147%	139%	0%	9.37%	0.02%	0.98%	0.00%	1.21%	8.75%	4.0	651	5.04
BD20	BLUE ^{M2040} _{C-CCU}	134%	126%	0%	8.55%	0.02%	0.55%	0.00%	1.18%	7.85%	3.6	576	5.00
BD20	GREY ^{M2040} _{C-ACR}	160%	151%	0%	10.24%	0.02%	0.92%	0.00%	1.19%	9.62%	4.4	704	5.07
BD20	GREEN ^{M2040} _{C-CCS}	92%	87%	0%	5.89%	0.02%	0.31%	0.00%	1.34%	6.04%	2.6	476	4.90
BD20	GREEN ^{M2040} _{C-CCU}	106%	100%	0%	6.78%	0.02%	0.22%	0.00%	1.25%	6.48%	2.9	487	4.93
BD20	YELLOW ^{M2040} _{NoCCU}	106%	100%	0%	6.76%	0.02%	0.25%	0.00%	1.33%	6.91%	2.9	529	4.93
BD20	GREY ^{M2020} _{C-ACR}	159%	151%	0%	10.18%	0.02%	0.97%	0.00%	1.22%	9.13%	4.4	722	5.07
BD20	GREY ^{M2040} _{C-ACR}	152%	144%	0%	9.71%	0.02%	0.91%	0.00%	1.19%	8.90%	4.1	646	5.05
BD20	YELLOW ^{M2020} _{C-ACR}	113%	108%	0%	7.25%	0.02%	0.38%	0.00%	1.40%	6.70%	3.2	639	4.95

BD20	<i>YELLOW</i> ^{M2040} _{C-ACR}	97%	92%	0%	6.22%	0.02%	0.25%	0.00%	1.32%	6.19%	2.7	471	4.91
BD20	<i>BLUE</i> ^{Wind} _{C-CCS}	145%	138%	0%	9.29%	0.02%	0.98%	0.00%	1.20%	8.33%	4.0	643	5.03
BD20	<i>BLUE</i> ^{Wind} _{C-CCU}	133%	126%	0%	8.49%	0.02%	0.55%	0.00%	1.17%	7.53%	3.6	570	5.00
BD20	<i>GREY</i> ^{Wind} _{NoCCU}	159%	150%	0%	10.14%	0.02%	0.91%	0.00%	1.19%	9.09%	4.3	694	5.07
BD20	<i>GREEN</i> ^{Wind} _{C-CCS}	90%	85%	0%	5.74%	0.02%	0.30%	0.00%	1.33%	5.25%	2.5	462	4.89
BD20	<i>GREEN</i> ^{Wind} _{C-CCU}	105%	99%	0%	6.69%	0.02%	0.21%	0.00%	1.24%	5.97%	2.9	478	4.93
BD20	<i>YELLOW</i> ^{Wind} _{NoCCU}	103%	97%	0%	6.59%	0.02%	0.24%	0.00%	1.31%	6.01%	2.8	513	4.92
BD20	<i>BLUE</i> ^{Wind} _{C-ACR}	151%	143%	0%	9.65%	0.02%	0.91%	0.00%	1.18%	8.59%	4.1	640	5.05
BD20	<i>YELLOW</i> ^{Wind} _{C-ACR}	95%	90%	0%	6.10%	0.02%	0.24%	0.00%	1.31%	5.51%	2.6	459	4.90

Rows in the table correspond to the scenario's biodiesel from microalgae. Acronyms for the scenario labels are as follows. BD20: Biodiesel 20% Vol; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; Wind: Electricity supplied from wind sources; DAC: CO₂ from Direct air capture; NGP: CO₂ from Natural gas power plant; CCU: Carbon capture and utilization of CO₂ from cogeneration by biomass combustion; CCS: Carbon capture and storage in a geological reservoir; C: cogeneration by biomass combustion; B: cogeneration by biogas combustion; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed; Grey: NGP and CCS or CCU is not performed; Yellow: DAC and CCS or CCU is not performed; NoCCU: LEA cogeneration is not considered (aCO₂: atmospheric CO₂ concentration EI: energy imbalance at the top of the atmosphere; SOD: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow- global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; CFP: Carbon footprint expressed in Gt CO₂eq; HH: Human health impacts expressed in DALYs; PBF: Planetary boundary footprint).

Table S28 Share of the global SOS, carbon footprint, Human health impacts, and PB footprint for biodiesel production occupied by global heavy-duty transport sector considering the current global electricity mix, 2040 sustainable electricity mix and wind electricity.

Fuel	Scenario	aCO ₂	EI	SOD	OA	P	N	LSC	FWU	CBI	CF	HH	PBF
		ppm	Wm ⁻²	DU	Ω _{arag}	Tg P	Tg N	% LSC	km ³	% BII loss	kg CO ₂ eq	DALY	
BD20	<i>BLUE</i> ^{M2020} _{B-CCS}	160%	152%	0%	10.24%	0.02%	0.99%	0.00%	1.22%	9.35%	4.5	727	5.07
BD20	<i>BLUE</i> ^{M2020} _{B-CCU}	152%	144%	0%	9.71%	0.02%	0.74%	0.00%	1.20%	8.82%	4.2	674	5.05
BD20	<i>GREY</i> ^{M2020} _{NoCCU}	173%	164%	0%	11.04%	0.02%	1.02%	0.00%	1.26%	10.02%	4.8	833	5.10
BD20	<i>GREEN</i> ^{M2020} _{B-CCS}	116%	110%	0%	7.40%	0.02%	0.39%	0.00%	1.40%	6.99%	3.3	648	4.96
BD20	<i>GREEN</i> ^{M2020} _{B-CCS}	119%	113%	0%	7.59%	0.02%	0.31%	0.00%	1.33%	7.05%	3.4	613	4.97
BD20	<i>YELLOW</i> ^{M2020} _{NoCCU}	127%	121%	0%	8.11%	0.02%	0.42%	0.00%	1.43%	7.58%	3.6	750	4.99
BD20	<i>BLUE</i> ^{M2040} _{B-CCS}	155%	147%	0%	9.93%	0.02%	0.95%	0.00%	1.20%	9.20%	4.3	677	5.06

BD20	<i>BLUE</i> ^{M2040} _{B-CCU}	148%	140%	0%	9.47%	0.02%	0.71%	0.00%	1.18%	8.70%	4.1	636	5.04
BD20	<i>GREY</i> ^{M2040} _{B-ACR}	160%	151%	0%	10.24%	0.02%	0.92%	0.00%	1.19%	9.62%	4.4	704	5.07
BD20	<i>GREEN</i> ^{M2040} _{B-CCS}	102%	97%	0%	6.54%	0.02%	0.28%	0.00%	1.33%	6.57%	2.9	507	4.92
BD20	<i>GREEN</i> ^{M2040} _{B-CCU}	109%	103%	0%	6.95%	0.02%	0.23%	0.00%	1.28%	6.74%	3.1	509	4.94
BD20	<i>YELLOW</i> ^{M2040} _{NoCCU}	106%	100%	0%	6.76%	0.02%	0.25%	0.00%	1.33%	6.91%	2.9	529	4.93
BD20	<i>GREY</i> ^{M2020} _{B-ACR}	172%	163%	0%	10.99%	0.02%	0.94%	0.00%	1.20%	9.95%	4.8	743	5.10
BD20	<i>GREY</i> ^{M2040} _{B-ACR}	168%	159%	0%	10.77%	0.02%	0.91%	0.00%	1.18%	9.84%	4.7	707	5.09
BD20	<i>YELLOW</i> ^{M2020} _{B-ACR}	126%	120%	0%	8.06%	0.02%	0.34%	0.00%	1.38%	7.52%	3.6	660	4.99
BD20	<i>YELLOW</i> ^{M2040} _{B-ACR}	114%	108%	0%	7.28%	0.02%	0.25%	0.00%	1.32%	7.14%	3.2	533	4.95
BD20	<i>BLUE</i> ^{Wind} _{B-CCS}	155%	147%	0%	9.89%	0.02%	0.95%	0.00%	1.19%	8.99%	4.3	674	5.06
BD20	<i>BLUE</i> ^{Wind} _{B-CCU}	148%	140%	0%	9.44%	0.02%	0.71%	0.00%	1.18%	8.54%	4.1	633	5.04
BD20	<i>GREY</i> ^{Wind} _{NoCCU}	159%	150%	0%	10.14%	0.02%	0.91%	0.00%	1.19%	9.09%	4.3	694	5.07
BD20	<i>GREEN</i> ^{Wind} _{B-CCS}	101%	95%	0%	6.43%	0.02%	0.28%	0.00%	1.32%	5.99%	2.9	497	4.92
BD20	<i>GREEN</i> ^{Wind} _{B-CCU}	107%	102%	0%	6.87%	0.02%	0.22%	0.00%	1.27%	6.32%	3.0	502	4.94
BD20	<i>YELLOW</i> ^{Wind} _{NoCCU}	103%	97%	0%	6.59%	0.02%	0.24%	0.00%	1.31%	6.01%	2.8	513	4.92
BD20	<i>BLUE</i> ^{Wind} _{B-ACR}	168%	159%	0%	10.74%	0.02%	0.91%	0.00%	1.18%	9.70%	4.7	705	5.09
BD20	<i>YELLOW</i> ^{Wind} _{B-ACR}	112%	107%	0%	7.19%	0.02%	0.24%	0.00%	1.31%	6.62%	3.2	523	4.95

Rows in the table correspond to the scenario's biodiesel from microalgae. Acronyms for the scenario labels are as follows. BD20: Biodiesel 20% Vol; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; Wind: Electricity supplied from wind sources; DAC: CO₂ from Direct air capture; NGP: CO₂ from Natural gas power plant; CCU: Carbon capture and utilization of CO₂ from cogeneration by biomass combustion; CCS: Carbon capture and storage in a geological reservoir; C: cogeneration by biomass combustion; B: cogeneration by biogas combustion; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed; Grey: NGP and CCS or CCU is not performed; Yellow: DAC and CCS or CCU is not performed; NoCCU: LEA cogeneration is not considered (aCO₂: atmospheric CO₂ concentration EI: energy imbalance at the top of the atmosphere; SOD: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow- global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; CFP: Carbon footprint expressed in Gt CO₂eq; HH: Human health impacts expressed in DALYs; PBF: Planetary boundary footprint).

Table S29. Share of the global SOS, carbon footprint, Human health impacts, and PB footprint for HTL production occupied by the global heavy-duty transport sector considering the current global electricity mix, 2040 sustainable electricity mix, and wind electricity.

	aCO ₂	EI	SOD	OA	P	N	LSC	FWU	CBI	CF	HH	PBFs
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Fuel	Scenario	ppm	Wm ⁻²	DU	Ω_{arag}	Tg P	Tg N	% LSC	km ³	% BII loss	kg CO ₂ eq	DALY	
HTL	BLUE ^{M2020} _{C-CCS}	206%	196%	0%	13.15%	0.04%	2.24%	0.00%	0.54%	15.95%	5.8	1116	5.20
HTL	BLUE ^{M2020} _{C-CCU}	179%	171%	0%	11.48%	0.04%	1.67%	0.00%	0.47%	14.27%	5.0	942	5.13
HTL	GREY ^{M2020} _{NoCCU}	241%	229%	0%	15.39%	0.04%	2.08%	0.00%	0.48%	17.73%	6.7	1153	5.29
HTL	GREEN ^{M2020} _{C-CCS}	121%	117%	0%	7.73%	0.04%	0.98%	0.00%	0.93%	11.52%	3.6	992	4.98
HTL	GREEN ^{M2020} _{C-CCS}	109%	106%	0%	6.98%	0.04%	0.75%	0.00%	0.76%	10.58%	3.2	826	4.95
HTL	YELLOW ^{M2020} _{NoCCU}	145%	140%	0%	9.29%	0.04%	0.82%	0.00%	0.87%	12.71%	4.2	995	5.04
HTL	BLUE ^{M2040} _{C-CCS}	178%	169%	0%	11.37%	0.04%	2.09%	0.00%	0.41%	15.08%	5.0	824	5.12
HTL	BLUE ^{M2040} _{C-CCU}	156%	148%	0%	9.97%	0.04%	1.54%	0.00%	0.36%	13.53%	4.3	695	5.07
HTL	GREY ^{M2040} _{C-ACR}	217%	206%	0%	13.88%	0.04%	1.95%	0.00%	0.37%	16.99%	6.0	906	5.22
HTL	GREEN ^{M2040} _{C-CCS}	74%	71%	0%	4.76%	0.04%	0.73%	0.00%	0.71%	10.07%	2.2	505	4.86
HTL	GREEN ^{M2040} _{C-CCU}	72%	68%	0%	4.59%	0.04%	0.54%	0.00%	0.58%	9.41%	2.1	435	4.85
HTL	YELLOW ^{M2040} _{NoCCU}	103%	98%	0%	6.59%	0.04%	0.59%	0.00%	0.67%	11.39%	2.9	553	4.93
HTL	BLUE ^{Wind} _{C-CCS}	174%	165%	0%	11.14%	0.04%	2.09%	0.00%	0.39%	13.84%	4.9	803	5.11
HTL	BLUE ^{Wind} _{C-CCU}	153%	145%	0%	9.77%	0.04%	1.54%	0.00%	0.34%	12.49%	4.2	677	5.06
HTL	GREY ^{Wind} _{NoCCU}	214%	203%	0%	13.68%	0.04%	1.95%	0.00%	0.35%	15.94%	5.9	888	5.22
HTL	GREEN ^{Wind} _{C-CCS}	69%	65%	0%	4.38%	0.04%	0.73%	0.00%	0.67%	8.00%	2.0	470	4.84
HTL	GREEN ^{Wind} _{C-CCS}	67%	64%	0%	4.29%	0.04%	0.54%	0.00%	0.55%	7.75%	1.9	407	4.84
HTL	YELLOW ^{Wind} _{NoCCU}	98%	93%	0%	6.24%	0.04%	0.60%	0.00%	0.63%	9.51%	2.8	521	4.91

Rows in the table correspond to the scenario's HTL biofuel from microalgae. Acronyms for the scenario labels are as follows. BD20: Biodiesel 20% Vol; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; Wind: Electricity supplied from wind sources; DAC: CO₂ from Direct air capture; NGP: CO₂ from Natural gas power plant; CCU: Carbon capture and utilization of CO₂ from cogeneration by biomass combustion; CCS: Carbon capture and storage in a geological reservoir; C: cogeneration by biomass combustion; B: cogeneration by biogas combustion; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed; Grey: NGP and CCS or CCU is not performed; Yellow: DAC and CCS or CCU is not performed; NoCCU: LEA cogeneration is not considered (aCO₂: atmospheric CO₂ concentration EI: energy imbalance at the top of the atmosphere; SOD: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow- global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; CFP: Carbon footprint expressed in Gt CO₂eq; HH: Human health impacts expressed in DALYs; PBF: Planetary boundary footprint).

Table S30. Share of the global SOS, carbon footprint, human health impacts, and PB footprint for HDO100 production occupied by the global heavy-duty transport sector considering the current global electricity mix, 2040 sustainable electricity mix, and wind electricity.

Fuel	Scenario	aCO ₂	EI	SOD	OA	P	N	LSC	FWU	CBI	CF	HH	PBFs
		ppm	Wm ⁻²	DU	Ω _{arag}	Tg P	Tg N	% LSC	km ³	% BII loss	kg CO ₂ eq	DALY	
HDO100	BLUE ^{M2020} _{C-CCS}	325%	310%	0%	20.76%	0.08%	4.54%	0.00%	5.46%	19.38%	9.2	1746	5.51
HDO100	BLUE ^{M2020} _{C-CCU}	255%	243%	0%	16.29%	0.08%	2.46%	0.00%	5.27%	14.91%	7.1	1295	5.32
HDO100	GREY ^{M2020} _{NoCCU}	400%	381%	0%	25.61%	0.08%	4.35%	0.00%	5.46%	23.85%	11.2	2125	5.70
HDO100	GREEN ^{M2020} _{C-CCS}	112%	111%	0%	7.15%	0.08%	1.76%	0.00%	6.28%	8.07%	3.7	1357	4.96
HDO100	GREEN ^{M2020} _{C-CCS}	146%	142%	0%	9.37%	0.08%	1.07%	0.00%	5.69%	9.15%	4.3	1095	5.05
HDO100	YELLOW ^{M2020} _{NoCCU}	188%	182%	0%	12.01%	0.08%	1.58%	0.00%	6.29%	12.54%	5.6	1736	5.16
HDO100	BLUE ^{M2040} _{C-CCS}	279%	264%	0%	17.82%	0.08%	4.17%	0.00%	5.22%	17.93%	7.8	1266	5.39
HDO100	BLUE ^{M2040} _{C-CCU}	219%	207%	0%	13.99%	0.08%	2.17%	0.00%	5.09%	13.77%	6.0	919	5.23
HDO100	GREY ^{M2040} _{C-ACR}	342%	322%	0%	21.86%	0.08%	3.89%	0.00%	5.17%	22.00%	9.4	1512	5.55
HDO100	GREEN ^{M2040} _{C-CCS}	25%	25%	0%	1.62%	0.08%	1.07%	0.00%	5.85%	5.34%	1.1	454	4.74
HDO100	GREEN ^{M2040} _{C-CCU}	90%	86%	0%	5.77%	0.08%	0.62%	0.00%	5.40%	7.38%	2.6	508	4.90
HDO100	YELLOW ^{M2040} _{NoCCU}	89%	83%	0%	5.66%	0.08%	0.78%	0.00%	5.79%	9.41%	2.7	700	4.90
HDO100	GREY ^{M2020} _{C-ACR}	338%	322%	0%	21.61%	0.08%	4.14%	0.00%	5.31%	19.74%	9.4	1608	5.54
HDO100	GREY ^{M2040} _{C-ACR}	303%	287%	0%	19.38%	0.08%	3.86%	0.00%	5.13%	18.64%	8.4	1243	5.45
HDO100	YELLOW ^{M2020} _{C-ACR}	125%	123%	0%	8.01%	0.08%	1.36%	0.00%	6.13%	8.43%	3.9	1219	4.99
HDO100	YELLOW ^{M2040} _{C-ACR}	50%	48%	0%	3.18%	0.08%	0.76%	0.00%	5.75%	6.05%	1.6	431	4.80
HDO100	BLUE ^{Wind} _{C-CCS}	273%	259%	0%	17.46%	0.08%	4.15%	0.00%	5.19%	15.98%	7.7	1231	5.37
HDO100	BLUE ^{Wind} _{C-CCU}	214%	203%	0%	13.70%	0.08%	2.16%	0.00%	5.07%	12.24%	5.9	892	5.22
HDO100	GREY ^{Wind} _{NoCCU}	335%	315%	0%	21.40%	0.08%	3.86%	0.00%	5.13%	19.51%	9.2	1468	5.53
HDO100	GREEN ^{Wind} _{C-CCS}	15%	15%	0%	0.94%	0.08%	1.03%	0.00%	5.79%	1.66%	0.8	388	4.70
HDO100	GREEN ^{Wind} _{C-CCU}	83%	79%	0%	5.33%	0.08%	0.60%	0.00%	5.36%	4.98%	2.4	465	4.88

HDO100	YELLOW ^{Wind} _{NoCCU}	76%	71%	0%	4.88%	0.08%	0.74%	0.00%	5.72%	5.19%	2.3	625	4.86
HDO100	BLUE ^{Wind} _{C-ACR}	299%	283%	0%	19.10%	0.08%	3.84%	0.00%	5.11%	17.15%	8.2	1217	5.44
HDO100	YELLOW ^{Wind} _{C-ACR}	40%	39%	0%	2.58%	0.08%	0.72%	0.00%	5.70%	2.83%	1.3	374	4.77

Rows in the table correspond to the scenario's biofuel from microalgae. Acronyms for the scenario labels are as follows. BD20: Biodiesel 20% Vol; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; Wind: Electricity supplied from wind sources; DAC: CO₂ from Direct air capture; NGP: CO₂ from Natural gas power plant; CCU: Carbon capture and utilization of CO₂ from cogeneration by biomass combustion; CCS: Carbon capture and storage in a geological reservoir; C: cogeneration by biomass combustion; B: cogeneration by biogas combustion; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed; Grey: NGP and CCS or CCU is not performed; Yellow: DAC and CCS or CCU is not performed; NoCCU: LEA cogeneration is not considered (aCO₂: atmospheric CO₂ concentration EI: energy imbalance at the top of the atmosphere; SOD: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow- global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; CFP: Carbon footprint expressed in Gt CO₂eq; HH: Human health impacts expressed in DALYs; PBF: Planetary boundary footprint).

Table S31. Share of the global SOS, carbon footprint, human health impacts, and PB footprint for HDO100 production occupied by the global heavy-duty transport sector considering the current global electricity mix, 2040 sustainable electricity mix, and wind electricity.

Fuel	Scenario	aCO ₂ ppm	EI Wm ⁻²	SOD DU	OA Ω _{arag}	P Tg P	N Tg N	LSC % LSC	FWU km ³	CBI % BII loss	CF kg CO ₂ eq	HH DALY	PBFs
HDO100	BLUE ^{M2020} _{B-CCS}	342%	326%	0%	21.90%	0.08%	4.21%	0.00%	5.29%	20.75%	9.9	1631	5.55
HDO100	BLUE ^{M2020} _{B-CCU}	304%	289%	0%	19.42%	0.08%	3.05%	0.00%	5.19%	18.28%	8.7	1383	5.45
HDO100	GREY ^{M2020} _{NoCCU}	479%	454%	0%	30.61%	0.08%	4.35%	0.00%	5.46%	28.20%	13.3	2376	5.91
HDO100	GREEN ^{M2020} _{B-CCS}	136%	134%	0%	8.71%	0.08%	1.43%	0.00%	6.11%	9.80%	4.5	1263	5.02
HDO100	GREEN ^{M2020} _{B-CCU}	150%	145%	0%	9.57%	0.08%	1.05%	0.00%	5.78%	10.08%	4.7	1100	5.06
HDO100	YELLOW ^{M2020} _{NoCCU}	266%	255%	0%	17.00%	0.08%	1.58%	0.00%	6.29%	16.89%	7.7	1988	5.36
HDO100	BLUE ^{M2040} _{B-CCS}	319%	303%	0%	20.42%	0.08%	4.03%	0.00%	5.17%	20.02%	9.2	1390	5.49
HDO100	BLUE ^{M2040} _{B-CCU}	286%	271%	0%	18.28%	0.08%	2.91%	0.00%	5.10%	17.71%	8.2	1196	5.41
HDO100	GREY ^{M2040} _{B-ACR}	420%	396%	0%	26.86%	0.08%	3.89%	0.00%	5.17%	26.35%	11.5	1764	5.75
HDO100	GREEN ^{M2040} _{B-CCS}	72%	70%	0%	4.64%	0.08%	0.92%	0.00%	5.79%	7.79%	2.6	599	4.86
HDO100	GREEN ^{M2040} _{B-CCU}	102%	98%	0%	6.56%	0.08%	0.67%	0.00%	5.54%	8.59%	3.3	608	4.93
HDO100	YELLOW ^{M2040} _{NoCCU}	167%	157%	0%	10.66%	0.08%	0.78%	0.00%	5.79%	13.75%	4.8	952	5.10
HDO100	GREY ^{M2020} _{B-ACR}	319%	304%	0%	20.39%	0.08%	3.99%	0.00%	5.20%	19.23%	9.2	1453	5.49

HDO100	<i>GREY</i> _{B-M2040-ACR}	302%	287%	0%	19.31%	0.08%	3.85%	0.00%	5.12%	18.69%	8.7	1277	5.45
HDO100	<i>YELLOW</i> _{B-M2020-ACR}	106%	105%	0%	6.79%	0.08%	1.21%	0.00%	6.03%	7.92%	3.6	1065	4.95
HDO100	<i>YELLOW</i> _{B-M2040-ACR}	49%	48%	0%	3.11%	0.08%	0.75%	0.00%	5.74%	6.10%	1.9	465	4.79
HDO100	<i>BLUE</i> _{B-Wind-CCS}	316%	301%	0%	20.24%	0.08%	4.01%	0.00%	5.16%	19.04%	9.1	1373	5.02
HDO100	<i>BLUE</i> _{B-Wind-CCU}	284%	269%	0%	18.14%	0.08%	2.90%	0.00%	5.09%	16.95%	8.1	1182	5.56
HDO100	<i>GREY</i> _{B-Wind-NoCCU}	413%	389%	0%	26.40%	0.08%	3.86%	0.00%	5.13%	23.85%	11.3	1720	5.60
HDO100	<i>GREEN</i> _{B-Wind-CCS}	65%	63%	0%	4.14%	0.08%	0.89%	0.00%	5.75%	5.08%	2.4	551	4.83
HDO100	<i>GREEN</i> _{B-Wind-CCU}	97%	93%	0%	6.19%	0.08%	0.65%	0.00%	5.51%	6.59%	3.1	573	4.92
HDO100	<i>YELLOW</i> _{B-Wind-NoCCU}	154%	145%	0%	9.88%	0.08%	0.74%	0.00%	5.72%	9.53%	4.4	877	5.06
HDO100	<i>BLUE</i> _{B-Wind-ACR}	300%	285%	0%	19.18%	0.08%	3.84%	0.00%	5.11%	17.98%	8.6	1264	5.15
HDO100	<i>YELLOW</i> _{B-Wind-ACR}	42%	41%	0%	2.66%	0.08%	0.72%	0.00%	5.70%	3.66%	1.7	422	5.31

Rows in the table correspond to the scenario's biofuel from microalgae. Acronyms for the scenario labels are as follows. BD20: Biodiesel 20% Vol; M2020: 2020 global electricity mix; M2040: Sustainable electricity mix for 2040; Wind: Electricity supplied from wind sources; DAC: CO₂ from Direct air capture; NGP: CO₂ from Natural gas power plant; CCU: Carbon capture and utilization of CO₂ from cogeneration by biomass combustion; CCS: Carbon capture and storage in a geological reservoir; C: cogeneration by biomass combustion; B: cogeneration by biogas combustion; Blue: NGP and either CCS or CCU is performed; Green: DAC and CCS or CCU is performed; Grey: NGP and CCS or CCU is not performed; Yellow: DAC and CCS or CCU is not performed; NoCCU: LEA cogeneration is not considered (aCO₂: atmospheric CO₂ concentration; EI: energy imbalance at the top of the atmosphere; SOD: stratospheric ozone depletion; OA: ocean acidification; P: biogeochemical phosphorus flow- global; N: biogeochemical nitrogen flow-global; LSC: land-system change-global; FWU: freshwater use, global; CBI: biosphere integrity; CFP: Carbon footprint expressed in Gt CO₂eq; HH: Human health impacts expressed in DALYs; PBF: Planetary boundary footprint).

3.2. Contributions to planetary boundaries

Figures S2 – S9 in this section we provide the breakdowns of every PB for the scenarios included in the main study with respect to the selected functional unit (FU), *i.e.*, the freight road transport (t km).

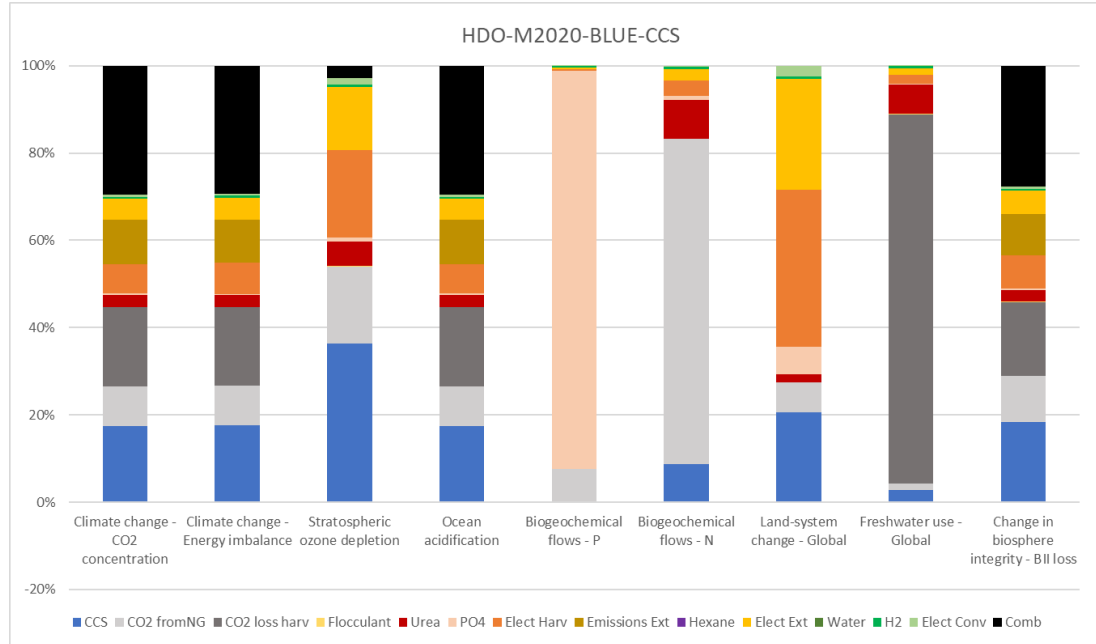


Figure S2. Breakdown of impact for planetary boundaries for HDO-BLUE^{M2020}_{CCS} (FU=1 t km).

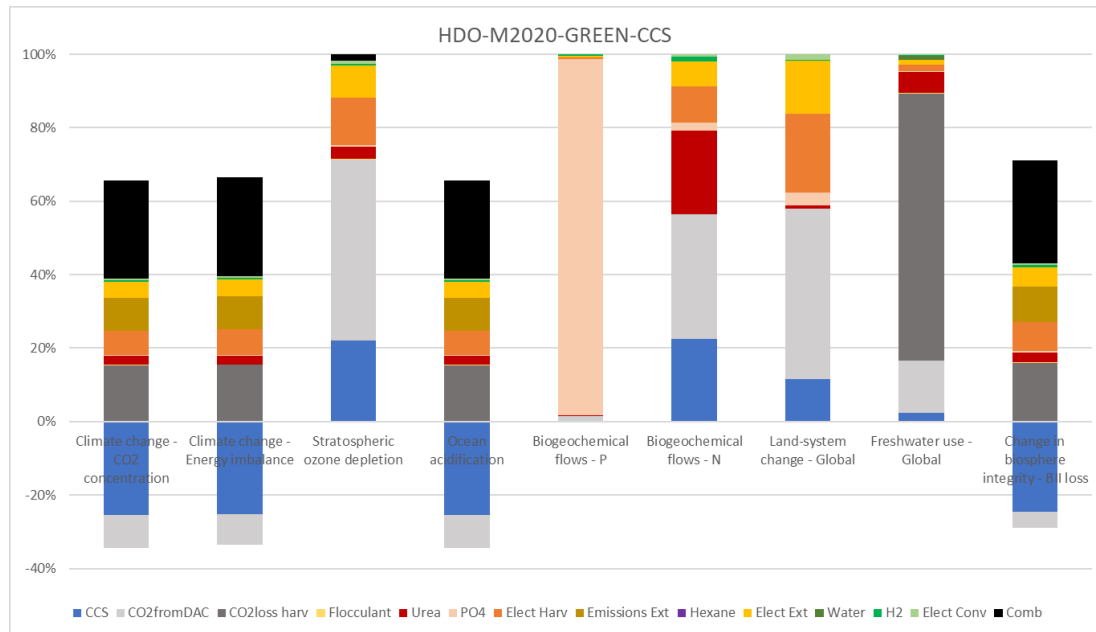


Figure S3. Breakdown of impact for planetary boundaries for HDO – GREEN^{M2020}_{CCS} (FU=1 t km).

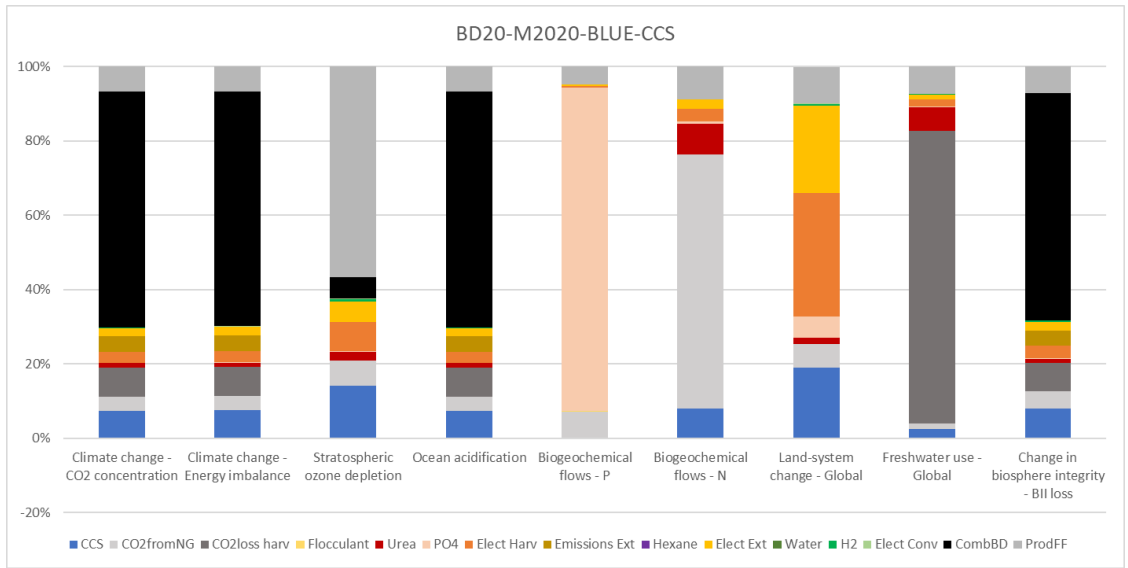


Figure S4. Breakdown of impact for planetary boundaries for BD20 – BLUE^{M2020}_{CCS} (FU=1 t km), the biodiesel fuel considered a blend with diesel 20% vol. BD.

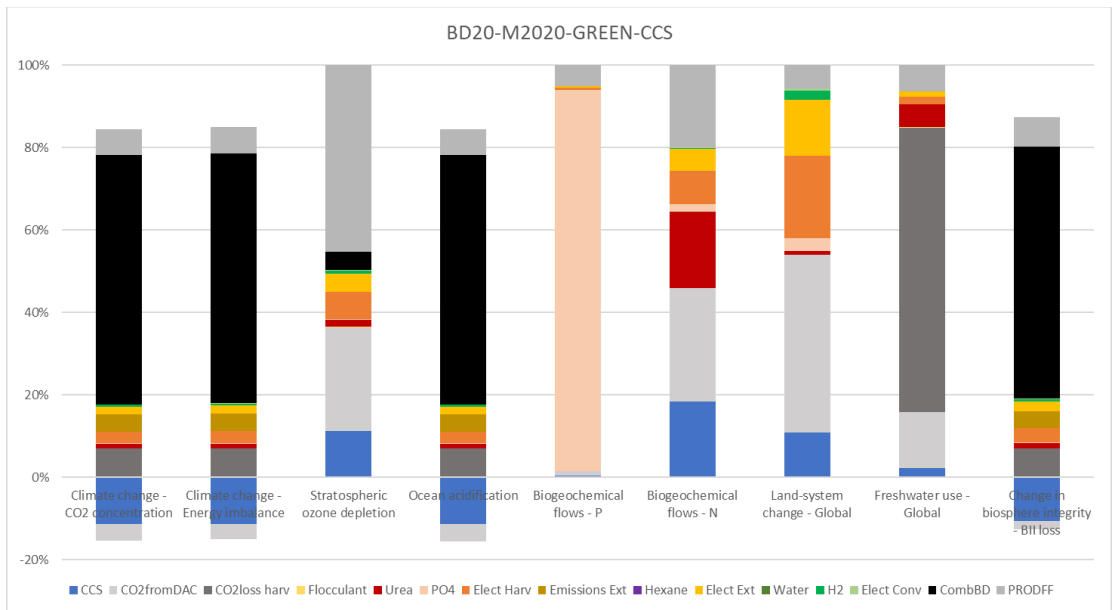


Figure S5. Breakdown of impact for planetary boundaries for BD20 – GREEN^{M2020}_{CCS} (FU=1 t km), the biodiesel fuel considered a blend with diesel 20% vol. BD.

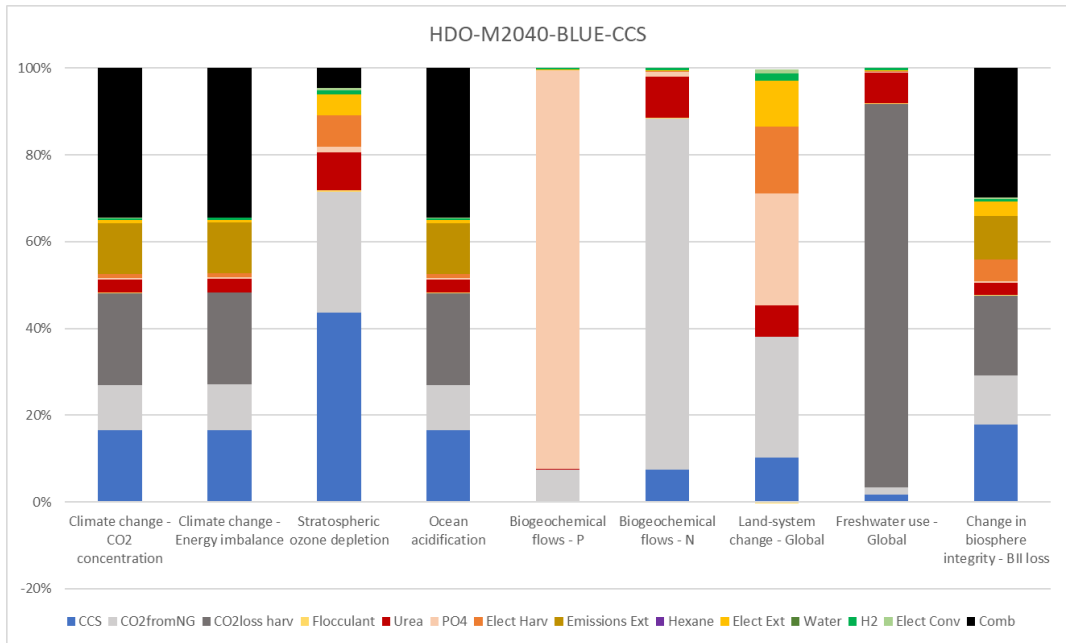


Figure S6. Breakdown of impact for planetary boundaries for HDO – BLUE^{M2040}_{CCS} (FU=1 t km)

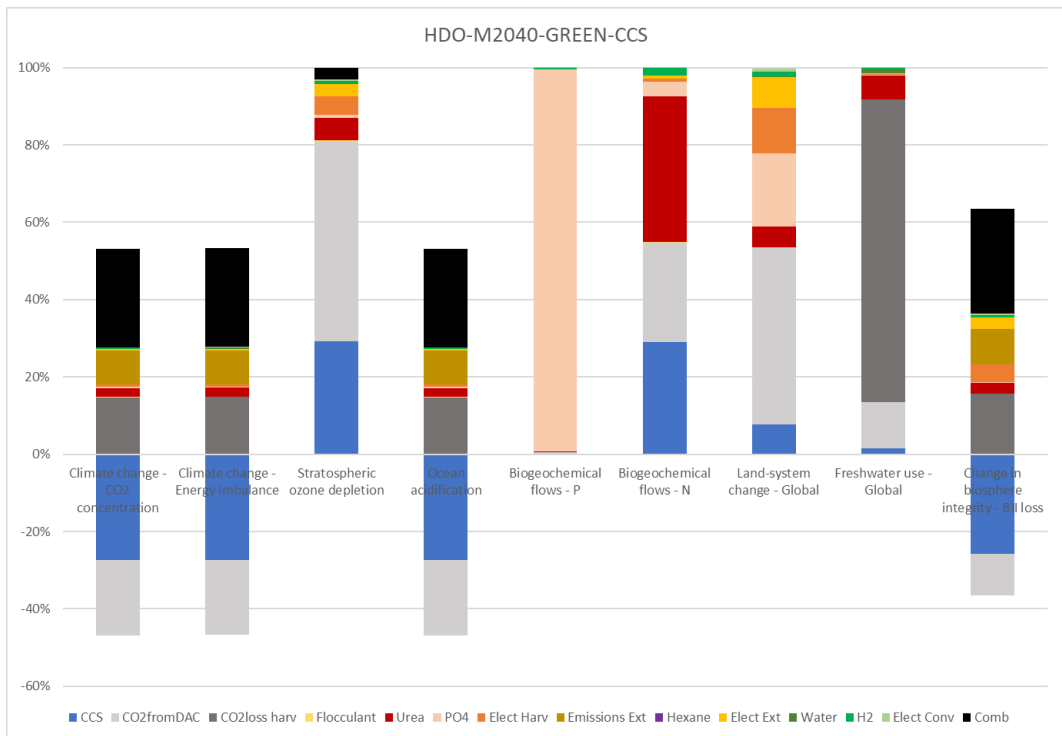


Figure S7. Breakdown of impact for planetary boundaries for HDO – GREEN^{M2040}_{CCS} (FU=1 t km).

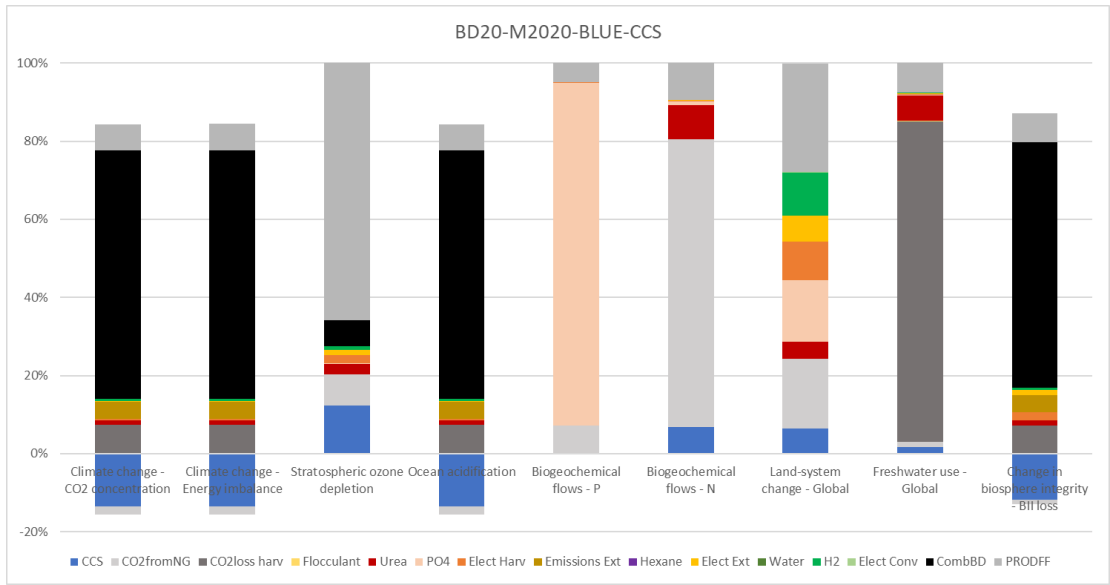


Figure S8. Breakdown of impact for planetary boundaries for BD20 – BLUE_{CCS}^{M2020} (FU=1 t km), the biodiesel fuel considered a blend with diesel 20% vol. BD.

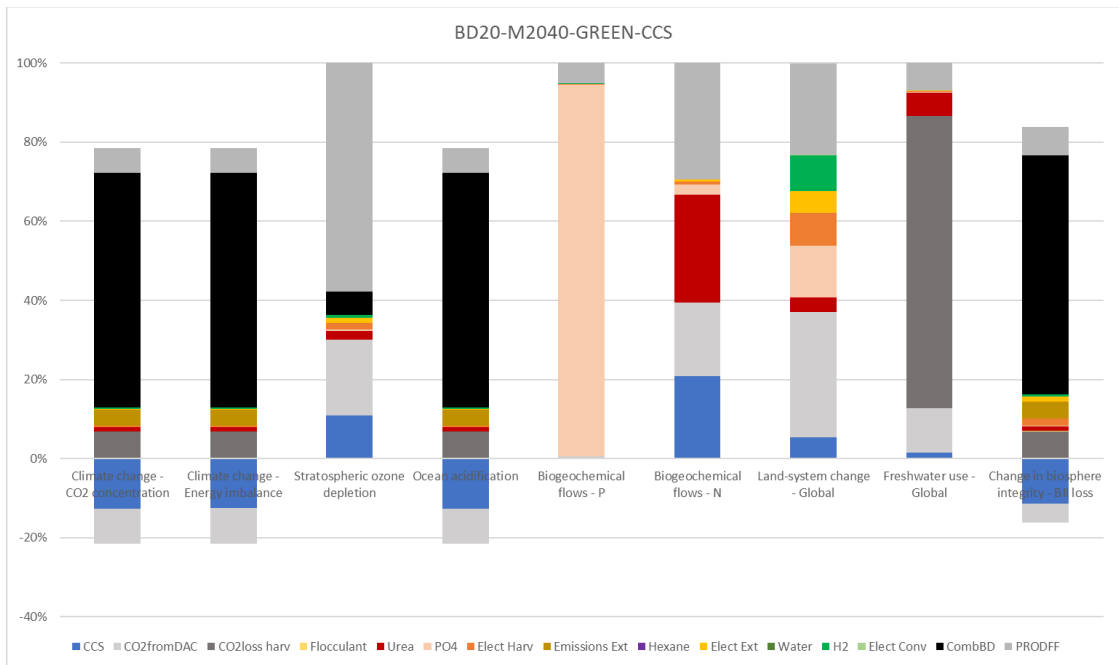


Figure S9. Breakdown of impact for planetary boundaries for BD20 – GREEN_{CCS}^{M2040} (FU=1 t km), the biodiesel fuel considered a blend with diesel 20% vol. BD.

Table S32. Technology readiness level of the main microalgae harvesting and extraction technologies

Stage	Technology	TRL	Reference
Cultivation	Open ponds	9	[152,169]
	Hanging bags	7	[152]
	Glass helical	7	[152]
	Horizontal Tubular	7	[152]
Harvesting	Flocculation	9	[152]
	Centrifugation	9	[152,170]
	Acoustic harvesting	6	[171]
Extraction	Dry extraction (Hexane)	9	[172]
	Wet extraction (Hexane, methanol)	9	[172]
	Sonification	4	[173]
	Supercritical CO ₂	3	[173]

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Supporting Information

The future of sustainable freight road transport: when and where will electric be more sustainable than biofuel powered trucks?

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This document is structured in 3 parts, section 1 provides details about the biofuel production process and their simulation using Aspen Plus, section 2 describes the techno-economic and brings additional information about the environmental assessment and section 3 presents additional information and the results not shown in the manuscript for the sake of shortness.

1. Overview of the biofuel production system

Several alternatives for biofuel production were simulated in this study. The first alternative, named biodiesel (BD), was an alkali-catalyzed process using vegetable oil as feedstock to produce biodiesel through transesterification reactions. The second option, named hydro-treated vegetable oil (HVO), was a hydrodeoxygenation process of vegetable oil to produce renewable diesel using catalysts. During these simulations, we have considered an annual treatment capacity of 42000 ton/year of vegetable oil, enabling the production of biofuel based on 8000 operating hours per year.

The two models were developed at Aspen Plus v12 based on previous studies [81,82,174,175]. In both models, triolein serves as a substitute vegetable oil feedstock. This compound is commonly selected to mimic vegetable oils due to its presence as a fatty acid constituent in a variety of vegetable oils, encompassing oils such as rapeseed, canola, olive, palm, and soybean oil [82,174,176,177].

1.1. Biodiesel production

Biodiesel production vegetable oil was modeled as 100% TGA triolein ($C_{57}H_{104}O_6$), considered as raw material to produce methyl oleate ($C_{19}H_{36}O_2$), obtained as the product of transesterification reactions (i.e., biodiesel).

The vegetable oil (i.e., triolein) reacts with excess methanol with the presence of a homogeneous alkali catalyst (sodium hydroxide) according to reaction 1.

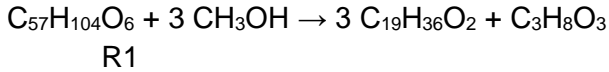


Figure S1 provides a flow diagram of the biodiesel production process. Triolein and methanol are mixed in 6:1 molar ratio and introduced into a reactor RSTOIC at 60°C and 4 bar, a reaction time of 60 min, and 95% conversion [82]. The non-random two liquid (NRTL) model was selected as a property package due to the presence of polar compounds [176].

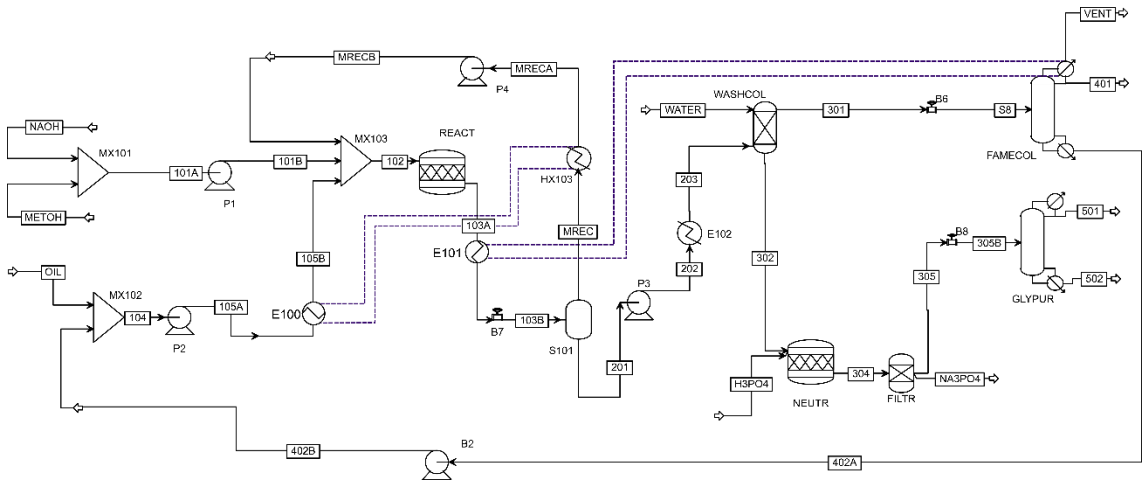


Figure S4. Production of biodiesel through transesterification of triolein using alkali catalyst.

The purple lines represent energy integration between equipment and/or streams.

After the transesterification reactor (REACT) the pressure is reduced to 0.1 bar. Subsequently, the product (103A) is directed into a flash separator (S101) at 80°C, where the vapor phase (MREC), mainly composed of methanol, is separated, and recycled into the process (MRECB).

The liquid phase, consisting of a mixture of fatty acid methyl esters (FAME), glycols, and water, is subjected to a washing column (WASHCOL) operating at atmospheric pressure. The objective is to separate the glycerol generated at the transesterification reactor (REACT). Subsequently, the stream (302), which encompasses the glycerol and water components, is directed to a neutralizing reactor (NEUTR), where a neutralization with H₃PO₄ solution. The final stream is purified in a distillation column (GLYPUR).

Stream 301, mainly composed of FAME or biodiesel (BD), is directed to a separation column (FAMECOL) that operates at 0.1 bar, where the gases are separated from the BD, and any unreacted triolein is recirculated to the

transesterification reactor (REACT). Both glycerol and biodiesel purification processes (GLYPUR and FAMECOL) are designed to achieve a purity level of 99.9 wt%.

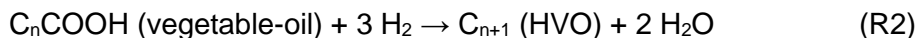
For the different separation processes considered in BD production, Universal Functional-Group Activity Coefficient Dortmund model (UNIFAC-DMD) is used. UNIFAC-DMD model has low deviations in temperature and vapor compositions when a mixture of water, fatty acids methyl oleate, and methanol are present [178,179].

1.2. Renewable diesel production

For this study, the production of HVO is based on previous models [174,180], which are composed of hydrogenation, separation, distillation, and pressure swing adsorption (PSA) to separate the hydrogen. It is assumed that hydrogen is externally supplied.

Figure S2 shows a flow diagram of the HVO process. The PSA was simulated as a “black box”, using data from Nordio et al. [181]

HVO is obtained from the reaction of hydrogen with vegetable oil, which is modeled as triolein, in a refinery-hydrotreating process. There are two main reactions: hydrodeoxygenation (R2) and decarboxylation (R3) [83].



A RSTOIC reactor was used due to the unavailability of a suitable kinetic model [174]. The oil is converted to gas (e.g., CO_2 , H_2 , propane, water, and HVO)

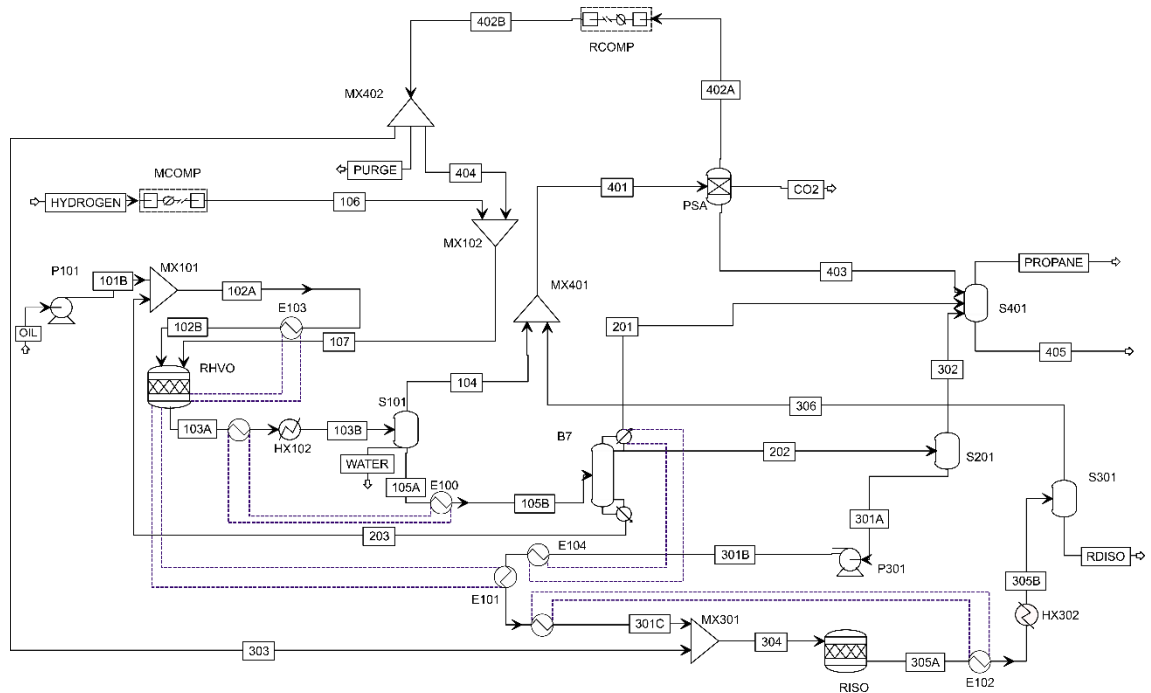
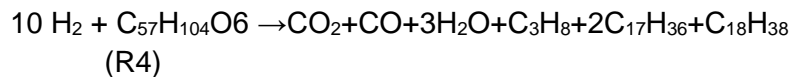


Figure S5 Production of HVO through hydrodeoxygenation of triolein.

The purple lines represent energy integration between equipment and/or streams.

HVO of vegetable oil was, therefore, represented by an RSTOIC reactor, which follows the following stoichiometry with a fractional conversion of 80% [174]:

In this reactor, RHVO, the triolein is hydrogenated. For simplicity the reaction is as follows [182]



The hydrogenation reaction conditions of triolein are 325°C and 32 bar where the amount of hydrogen supplied to the HVO reactor is 1.5 times the stoichiometric requirement and a reaction time of 60 min. In this process the vegetable oil is preheated at 90°C and pumped into the reactor (RHVO).

The exit steam from reactor RSTO (RHVO) was further cooled to 27°C before entering a three-phase flash drum S101 (27 °C and 12.8 bar). In this flash drum, water and hydrogen were separated from the hydrocarbons and CO₂. The gaseous stream containing hydrogen, CO₂, and light hydrocarbons is sent to a PSA to be separated (401). The separated flow of hydrogen is compressed to 32 bar and recycled to the reactor (RHVO) after a nominal purge (5%).

For the separation of hydrogen, CO₂ and light hydrocarbons we used the work of Nordio et al. which takes into account an electricity consumption of 20 kWh/kg of

hydrogen and a cost of 5.88 USD\$/kg of hydrogen that will be considered in the following analysis [181]. The hydrocarbon stream (105A) is heated to 90°C and sent to a separation column where the unreacted triolein is removed and sent back to the reactor RSTO while the stream containing the hydrocarbons is sent to a flash drum at 1 bar and 35°C where the propane is separated.

The stream containing the HVO (301) is pumped and heated to 32 bar and 234 °C before being introduced into the isomerization reactor (RISO) [183,184], where, the linear paraffin is generally hydro-isomerized to the branched paraffin to match the specifications with fossil-diesel. For a final separation the stream 305B is sent to a flash drum (RDISO) to separate the light hydrocarbons produced during the isomerization process.

The operating conditions, equipment sizing, and number of distillation stages were optimized using sensitivity analysis. On top of that, energy integration was included.

1.3. Simulations summary

The following section shows the equipment used for both processes, Table S1 summarizes the equipment involved in the simulations performed for BD and HVO production.

Table S3 Equipment summary

Equipment	Description	Specification		
Reactors		Volume [m ³]	Design temperature [°C]	Pressure [bar]
NEUTR	Agitated reactor	0.375	85	1
REACT	Agitated reactor	7.78	60	4
Vessels		Diameter [m]	Length [m]	Pressure [bar]
S101	Vertical	1.0668	2.74	0.1
FILTR	Vertical	0.91	1.22	1
Towers			Tower MOC	Pressure [bar]
FAMECOL	14 sieve trays		CS	0.1
GLYPUR	12 sieve trays		CS	0.4
WASHCOL	10 sieve trays		CS	1
Pump		Discharge pressure [bar]	Power [kWh]	[m ³ /h]
P1		4	0.2	0.72
B2/p5		1	0.04	0.54
P2		4	1.5	6.9
P3		1.1	0.6	8.3
P4		4	0.18	0.66

Exchangers		Pressure [bar]	Load [kJ/hr]	Area [m ²]
HX103	Cooler	0.1	561000	11.38
GLYPUR	Condenser	0.4	668900	2.341
FAMECOL	Condenser	0.1	2084000	3.324
GLYPUR	Reboiler	0.4	904400	6.37
FAMECOL	Reboiler	0.1	4199000	41.85
E100	Heat exchanger	4	786800	6.399
E101	Heat exchanger	4	105700	8.615
REACT	Cooler	4	342000	13.45

Process simulation is used to quantify the energy and mass flows required to produce the biofuels as shown in Table S2, which will later be used to evaluate their economic and environmental performance.

Table S4 Material and energy data for biofuel production

Raw material	RD	BD
Inlet		
Canola oil [kg/h]	5250	5250
Hydrogen [kg/h]	155.9	
Catalyst [kg/h]	0.01389	kg/h
Water for process [kg/h]		262.5
Sodium hydroxide [kg/h]		105
Methanol [kg/h]		577.5
H ₃ PO ₄ [kg/h]		262.5
Utilities		
Cooling water [m ³ /h]	148.8	95.2
Heat (Fuel gas) [MJ/h]	8890	5982
Medium pressure steam [MJ/h]	38.94	0.113
Electricity [kWh]	1319.6	2.5
Outlet (products and byproducts)		
FAME [kg/h]	0	5273.4
HVO [kg/h]	4347.8	
Propane [kg/h]	287.56	0
Glycerol [kg/h]		0
		545.6

1.4. Techno-economic and environmental assessment

The following section summarizes the methodology applied to assess the economic and environmental performance of conventional biodiesel and HVO by transesterification and hydrodeoxygenation, respectively. The total cost follows the methodology proposed by Towler and Sinnott [185], as a basis for the techno-

economic calculation. The environmental performance is evaluated under the LCA principles [27,89].

The total capital investment is an estimate for establishing a plant and refers to the total cost of designing, constructing, and installing a plant. The total capital investment is calculated as the sum of the inside battery limits (ISBL), the outside battery limits (OSBL), the engineering cost, and the contingency cost.

The ISBL cost represents the cost of the plant itself, the modifications and improvements that must be made to the site infrastructure OSBL, engineering and construction costs, and the contingency charges.

ISBL plant cost includes the cost of procuring and installing all the process equipment that makes up the new plant, all the major process equipment, bulk items (i.e., piping, valves, wiring, instruments, structures, etc.), civil works, installation labor and supervision; all of the above elements are also known as direct costs. In addition to the direct field costs, there will be indirect field costs including temporary construction costs, construction insurance, labor benefits, patent fees, etc.

OSBL plant cost is the off-site cost and includes the cost associated with the necessary modification and improvements made on the site of the studied plan infrastructure. For typical chemicals projects, offsite costs are usually between 20% and 50% of ISBL cost in this study and 29% will be used as an estimate. [185]

The equipment cost is estimated considering the purchased cost of major equipment based, Table S3 presents a summary of these equipments and their specifications. The specifications are determined based on data retrieved from the simulation, which additionally provides the material and energy balances for BD and HVO production. Details on the material of construction and the installation cost are added using the installation factors mentioned by Sinnott and Towler [185].

Finally, the cost escalation index, known as the Chemical Engineering Plant Cost Index (CEPCI), is used to obtain the final total capital plant cost. The CEPCI value published in the year 2015 is 532,9, and for the year 2021 is 720. The total capital investment calculated for both production plants is shown in Table S3.

Table S5 Capital investment cost for BD and HVO diesel production

Item	HVO [MillionUSD\$]	BD [MillionUSD\$]	Notes
Purchased equipment cost	3.50	0.86	
Total ISBL cost 2015	12.80	3.31	CEPI 2015
Total ISBL cost 2021	17.30	4.48	CEPI 2021
Warehouse	0.70	0.18	4% of the ISBL
Site development	1.60	0.40	9% of the ISBL
Additional piping	0.80	0.20	4.5% of the ISBL
Total direct cost	20.30	5.26	
Total indirect cost	5.00	1.30	29% of the ISBL
Total capital investment (TCI)	25.40	6.56	

The incorporation of OPEX (Operational Expenditure) and CAPEX (Capital Expenditure) in the determination of total product cost provides a comprehensive assessment associated with producing a product.

CAPEX refers to the initial investments made in acquiring assets such as equipment, machinery, facilities, and infrastructure required for the production process. OPEX includes the ongoing costs associated with running and maintaining the production process. It covers expenses such as labor, raw materials, energy, maintenance, repairs, and other operational costs.

The yearly cost for these investments is based on the total capital investment over the lifetime named as CAPEX. The annual CAPEX was calculated considering 15 years lifetime and 6% interest. The annual CAPEX was calculated based in the following equation [186]:

$$R = \frac{P}{\frac{(1+i)^n - 1}{i(1+i)^n}} \quad (\text{Eq S1})$$

Where:

R = annual equivalent cost (USD\$).

P = total capital investment present value (USD\$).

i = interest rate.

n = number of years.

As part of the OPEX we have the fixed cost and the variable cost of production. The variables that will be estimated and their relevant assumptions in the calculations are summarized in Table S4.

Table S6. Total fixed cost in Million USD\$/year.

	HVO	BD	
Maintenance	0.52	0.13	3% of ISBL
Labor cost	1.00	1.00	5 workes 4 turns
Supervision cost	0.25	0.25	25% of labor cost
Insurance and local taxes	0.35	0.09	2% of ISBL
Total fixed cost	2.12	1.47	

In the fixed cost, labor cost is one of the main variables. According to Towler and Sinnott [185], the plant requires 4 shifts and 5 operators per shift with a salary of 50000 USD\$ per year.

On the other hand, the variable production costs considered in this study are composed of the cost of raw materials (Table S6) and utilities required (Table S7).

Table S7. Prices of raw materials and utilities used in this study.

	Price	Units	Reference
Prices of raw materials			
Soybean oil	1612	USD\$/Mt	[187]
Canola oil	1024	USD\$/Mt	[187]
Palm oil	891	USD\$/Mt	[187]
Microalgae oil	2371	USD\$/Mt	[80,160]
Hydrogen	3.3	US\$/kg	[188]
Water for process	0.0003	US\$/kg	[32]
Sodium hydroxide	0.311	US\$/kg	[32]
Methanol	0.182	US\$/kg	[32]
H ₃ PO ₄	0.744	US\$/kg	[32]
Glycerol	0.5	US\$/kg	[32]
Propane	0.267	US\$/kg	[32]
Prices of utilities			
Cooling water	0.013	USD\$/m3	[185]
Medium pressure steam			[32]
Low pressure steam	0.014	USD\$/MJ	[32]
Fired heat	0.014	USD\$/MJ	[32]
Electricity	0.097	USD\$/kWh	[32]
Diesel	*0.76	USD\$/l	[189]

*In a further step, the price of the trucks will be taken into account to determine the cost per tkm transported, this price was obtained from a review and considers the values of USD \$80,000 USD for ICT and USD \$170,000 USD for BET [190] The operational costs of ICTs are estimated at around 0.010 USD\$/tkm, while, for BETs, it can be around 0.018 USD\$/tkm, considering 710000 km traveled over the lifetime of both trucks .

For the comparative value of the diesel price we took as a reference value 1.24 USD\$/L of which 39% corresponding to distribution and tax costs were reduced since these are not considered when determining the total costs of biofuel production [189].

Table S7 provides the total biofuel production costs based on the four types of biomass studied. The sum of CAPEX and OPEX divided by the annual fuel production shows the total production cost per kilogram of fuel. In this cost the taxes and profit margins are not considered.

Table S8 Production fuel cost

	Soybean		Palm		Canola		Microalgae	
	HVO	BD	HVO	BD	HVO	BD	HVO	BD
Annual CAPEX	2.98	0.68	2.98	0.68	2.98	0.68	2.98	0.68
Total fixed cost [Million US\$/year]	2.12	1.25	2.12	1.25	2.12	1.25	2.12	1.25
Total variable cost [Million US\$/year]	74.07	68.80	43.79	38.52	49.26	44.11	105.91	100.69
Total annual cost [Million US\$/year]	79.17	70.73	48.88	40.45	54.35	46.04	111.00	102.62
Byproduct revenue								
Propane [Million US\$/year]	0.61		0.61		0.61		0.61	
Glycerol [Million US\$/year]		2.18		2.18		2.18		2.18
Total [Million US\$/year]	78.55	68.54	48.27	38.26	53.74	43.85	110.39	100.43
Unit production cost [US\$/kg]	2.26	1.62	1.39	0.91	1.55	1.04	3.17	2.38
Unit production cost [US\$/kg]	1.64	1.18	1.64	0.66	1.64	0.76	1.64	1.73

1.5. Global temperature increases calculation

In the cases of the GWP indicator, the amount of GHG emissions avoided can be translated into an equivalent avoided temperature increase by using the relationship proposed by Hare and Meinshausen [191] (Eq. (S2)). According to this equation, a temperature change of about 0.16 °C is expected for every additional 100 Gt of fossil CO₂ accumulated in the atmosphere.

$$\Delta T_{g,p,s}^{t_1,t_2} = \frac{0.16^\circ\text{C}}{100 \text{ GtCO}_2} \cdot \sum_{t=t_1}^{t_2} \Delta IMP_{b=GWP,g,p,s,t} \quad \forall g, p, s \quad \text{Eq. (S2)}$$

Here, the cumulative CO₂ emissions avoided in every year t in region g under IAM scenario s , as given by variable $\Delta IMP_{b=GWP,g,p,s,t}$, is obtained by the difference between the expected emissions under the dICTs scenario (i.e., $p = dICTs$) and any sustainable scenario the scenario proposed (i.e., $p = \{bioICT, BETs\}$ see Eq. 5 in the main manuscript). Since calculations were only performed for 12 specific years (see section 2.2 in the main manuscript), annual emissions for intermediate years were determined by linear interpolation.

2. Life cycle inventories information

Table S8 and Table S9 provide the different life cycle inventories considered for biofuel production based on the process simulation performed and using the Ecoinvent v3.8 database.

Table S9. LCI of the foreground system for biodiesel production.

Process: BD production from vegetable oil			
	Ecoinvent entry	Description	Amount
Inputs:	Market for heat, from steam, in chemical industry, RoW	Heat for utilities [MJ]	1.13
	Electricity mix	Electricity for utilities [kWh]	0.0005

		*Vegetable oil [kg]	0.99
	Market for methanol, GLO	Methanol [kg]	0.11
	Market for phosphoric acid, industrial grade, without water, in 85% solution state, GLO	Acid for neutralization [kg]	0.05
	Market for sodium hydroxide, without water, in 50% solution state, GLO	Alkali catalyst [kg]	0.02
	Market group for tap water, GLO	Water for washing column [kg]	0.05
Outputs:		Glycerol [kg]	0.1
		BD fuel [kg]	1

Table S10. LCI of the foreground system for HVO fuel production

Process: HVO production from vegetable oil			
	Ecoinvent entry	Description	Amount
Inputs:			
	Market for hydrogen, gaseous, GLO	Hydrogen input [kg]	0.035
	Market for heat, from steam, in chemical industry, RoW	Heat for utilities [MJ]	2
		*Vegetable oil [kg]	1.3
	Electricity mix	Electricity for utilities [kWh]	0.3
Outputs:			
		Propane [kg]	0.065

HVO fuel [kg] 1

* The inventories name as vegetable oil, vary in the different scenarios studied and may be; market for rape oil, crude RoW; market for soybean oil, crude GLO; market for palm oil, crude GLO; microalgae oil considering direct air capture and carbon capture storage [80].

For the building of the LCIs concerning the use of trucks, the existing inventories in the *Premise* database are used as a basis[69]. Table TS11 summarizes the variation of these parameters in the time interval 2020 to 2100, which is studied in this work.

Table S11 BET and ICT parameter changes from 2020 to 2100

		2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
Km over lifetime	km	71000	71000	71000	71000	71000	71000	71000	71000	71000	71000	71000	71000	71000
Yearly mileage	km/year	0	0	0	0	0	0	0	0	0	0	0	0	0
Autonomy on a full tank/battery	km	10700	10700	10700	10700	10700	10700	10700	10700	10700	10700	10700	10700	10700
Payload	kg	0	0	0	0	0	0	0	0	0	0	0	0	0
	kg	800	800	800	800	800	800	800	800	800	800	800	800	800
	kg	13800	13800	13800	13800	13800	13800	13800	13800	13800	13800	13800	13800	13800
ICT Tank-to-wheel energy consumption	kJ/tkm													
	kJ/tkm	852	723	633	623	613	592	572	572	572	572	572	572	572
Tank-to-wheel efficiency	%													
	%	0.31	0.31	0.33	0.33	0.33	0.33	0.34	0.34	0.34	0.34	0.34	0.34	0.34
ICT weight	kg													
	kg	14891	13660	12571	12030	11501	10986	10471	10471	10471	10471	10471	10471	10471

BET

Battery-to-wheel energy consumption	kJ/tkm	480	386	316	297	281	265	249	249	249	249	249	249	249
Battery-to-wheel efficiency		0.72	0.72	0.74	0.74	0.76	0.76	0.77	0.77	0.77	0.77	0.77	0.77	0.77
BET weight	kg	22794	22794	18712	16708	15179	13905	12700	12700	12700	12700	12700	12700	12700
Batter weight	kg	7888	7888	5380	3898	2900	2173	1640	1640	1640	1640	1640	1640	1640

The "Electricity" activity differs depending on the region and the year. Table S12 to Table S24 provides a thorough breakdown of the various electricity scenario options. The different mixes shown were constructed from the information obtained from IISA for the two scenarios studied, namely "Current policies" (CP) and "NetZero policies" (NZ) [74,192].

Table S12. Breakdown of the European Union and UK region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	8%	13%	20%	24%	27%	28%	27%	24%	24%	26%	26%	27%
Hydro	13%	13%	11%	10%	10%	9%	9%	9%	8%	7%	7%	6%
Wind	15%	24%	31%	37%	43%	49%	55%	63%	65%	64%	64%	63%
Gas	5%	5%	4%	3%	2%	1%	0%	0%	0%	0%	0%	0%
Coal	20%	12%	8%	5%	3%	1%	0%	0%	0%	0%	0%	0%
Geothermal	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%
Nuclear	31%	26%	20%	15%	11%	8%	5%	1%	0%	0%	0%	0%
Oil	2%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	5%	5%	5%	4%	4%	3%	3%	3%	2%	2%	3%	3%
NetZero												
Solar	8%	17%	26%	29%	31%	31%	30%	27%	27%	29%	29%	29%
Hydro	13%	13%	11%	9%	8%	8%	7%	6%	5%	5%	4%	4%
Wind	15%	29%	38%	44%	48%	52%	56%	64%	66%	64%	64%	64%
Gas	5%	5%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	20%	2%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	1%	1%	1%	1%	1%	1%	1%	1%	1%	0%	0%	0%
Nuclear	31%	27%	19%	13%	9%	6%	4%	1%	0%	0%	0%	0%
Oil	2%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	5%	5%	5%	4%	3%	3%	2%	2%	1%	1%	1%	1%

Table S13. Breakdown of the China region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	10%	15%	20%	23%	26%	28%	30%	31%	33%	34%	35%
Hydro	18%	21%	19%	17%	16%	16%	17%	18%	19%	20%	21%	22%
Wind	5%	9%	12%	15%	17%	18%	21%	28%	33%	35%	37%	37%
Gas	1%	2%	3%	4%	4%	4%	3%	3%	1%	0%	0%	0%
Coal	65%	50%	41%	35%	30%	25%	19%	10%	6%	3%	1%	1%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	6%	7%	8%	9%	9%	10%	11%	11%	9%	6%	4%	1%
Oil	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	1%	1%	1%	1%	1%	1%	1%	1%	2%	3%	4%
NetZero												
Solar	3%	20%	33%	36%	37%	38%	38%	35%	34%	39%	42%	41%
Hydro	18%	24%	19%	16%	14%	14%	14%	16%	16%	17%	18%	17%
Wind	5%	18%	28%	33%	35%	35%	34%	35%	37%	35%	35%	39%
Gas	1%	5%	6%	4%	2%	0%	0%	0%	0%	0%	0%	0%

Richard Emmanuel Cabrera Jiménez

Coal	65%	23%	3%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	6%	9%	10%	10%	11%	12%	12%	13%	11%	8%	4%	2%
Oil	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	2%

Table S14. Breakdown of the USA electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	9%	20%	28%	34%	38%	40%	41%	40%	42%	43%	44%
Hydro	7%	7%	7%	7%	7%	7%	7%	6%	6%	6%	5%	5%
Wind	8%	14%	21%	28%	32%	37%	41%	46%	50%	48%	49%	48%
Gas	33%	29%	22%	19%	14%	10%	6%	3%	0%	0%	0%	0%
Coal	26%	21%	13%	6%	3%	1%	0%	0%	0%	0%	0%	0%
Geothermal	2%	2%	2%	2%	2%	2%	2%	2%	2%	2%	1%	1%
Nuclear	17%	15%	12%	9%	7%	5%	3%	1%	0%	0%	0%	0%
Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	2%	2%	2%	1%	1%	1%	1%	2%	2%	2%	2%
NetZero												
Solar	3%	17%	33%	40%	42%	42%	43%	42%	42%	45%	46%	46%
Hydro	7%	8%	7%	6%	6%	6%	6%	6%	5%	4%	4%	3%
Wind	8%	21%	34%	40%	42%	43%	45%	47%	50%	48%	48%	48%
Gas	33%	31%	11%	2%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	26%	2%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	2%	2%	2%	2%	1%	1%	1%	1%	1%	1%	1%	1%
Nuclear	17%	16%	11%	8%	6%	4%	3%	1%	0%	0%	0%	0%
Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	2%	3%	3%	3%	3%	3%	3%	2%	2%	1%	1%

Table S15. Breakdown of the Latin America region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	2%	8%	17%	24%	27%	29%	29%	29%	29%	31%	31%	31%
Hydro	59%	55%	47%	39%	34%	30%	27%	23%	21%	20%	20%	19%
Wind	6%	12%	18%	24%	28%	33%	36%	43%	46%	44%	43%	42%
Gas	10%	7%	6%	5%	4%	3%	2%	1%	1%	0%	0%	0%
Coal	7%	5%	3%	2%	1%	1%	1%	0%	0%	0%	0%	0%
Geothermal	1%	2%	2%	2%	1%	1%	1%	1%	1%	1%	1%	1%
Nuclear	2%	2%	2%	1%	1%	1%	0%	0%	0%	0%	0%	0%
Oil	7%	4%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	6%	5%	5%	4%	3%	3%	3%	2%	3%	4%	6%	6%
NetZero												

Richard Emmanuel Cabrera Jiménez

Solar	2%	12%	24%	30%	33%	33%	33%	32%	32%	34%	35%	36%
Hydro	59%	58%	43%	34%	29%	25%	23%	20%	18%	17%	17%	16%
Wind	6%	14%	22%	29%	33%	37%	40%	44%	47%	47%	47%	46%
Gas	10%	3%	2%	1%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	7%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	1%	2%	2%	2%	1%	1%	1%	1%	1%	1%	1%	1%
Nuclear	2%	2%	1%	1%	1%	0%	0%	0%	0%	0%	0%	0%
Oil	7%	4%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	6%	6%	5%	4%	4%	3%	3%	3%	2%	2%	1%	1%

Table S16. Breakdown of the Canada, Australia and New Zealand region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	4%	8%	13%	17%	20%	21%	21%	20%	20%	22%	23%	26%
Hydro	48%	51%	50%	47%	46%	45%	44%	42%	41%	39%	36%	31%
Wind	6%	11%	13%	17%	20%	24%	27%	32%	35%	36%	39%	41%
Gas	5%	4%	3%	2%	1%	0%	0%	0%	0%	0%	0%	0%
Coal	21%	10%	6%	4%	3%	0%	0%	0%	0%	0%	0%	0%
Geothermal	3%	4%	4%	4%	4%	4%	3%	2%	1%	0%	0%	0%
Nuclear	10%	9%	8%	6%	4%	3%	2%	1%	0%	0%	0%	0%
Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	2%	3%	3%	3%	2%	2%	3%	2%	2%	2%	2%
NetZero												
Solar	4%	11%	20%	24%	25%	25%	24%	22%	23%	25%	27%	29%
Hydro	48%	55%	49%	44%	40%	39%	38%	38%	36%	33%	29%	25%
Wind	6%	13%	16%	21%	25%	29%	32%	37%	39%	40%	42%	45%
Gas	5%	3%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	21%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	3%	4%	4%	4%	3%	3%	2%	1%	0%	0%	0%	0%
Nuclear	10%	10%	8%	5%	4%	3%	2%	1%	0%	0%	0%	0%
Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	3%	3%	3%	2%	2%	2%	2%	2%	2%	1%	1%

Table S17. Breakdown of the Africa region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	7%	12%	18%	22%	28%	32%	38%	41%	43%	44%	45%
Hydro	35%	39%	36%	30%	26%	22%	18%	12%	9%	7%	5%	4%
Wind	2%	6%	12%	18%	22%	28%	33%	41%	45%	48%	49%	48%
Gas	9%	13%	19%	23%	23%	20%	15%	8%	3%	1%	0%	0%
Coal	45%	29%	16%	8%	4%	1%	0%	1%	1%	2%	2%	3%
Geothermal	2%	3%	2%	2%	1%	1%	1%	0%	0%	0%	0%	0%
Nuclear	3%	2%	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%
Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	1%	1%	1%	1%	1%	0%	0%	0%	0%	0%	0%
NetZero												
Solar	3%	11%	24%	33%	37%	40%	41%	44%	44%	46%	49%	52%
Hydro	35%	42%	37%	30%	22%	17%	13%	9%	6%	5%	4%	3%
Wind	2%	9%	20%	32%	38%	41%	44%	46%	48%	48%	46%	44%
Gas	9%	9%	6%	2%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	45%	21%	7%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	2%	3%	2%	1%	1%	1%	0%	0%	0%	0%	0%	0%
Nuclear	3%	2%	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%

Richard Emmanuel Cabrera Jiménez

Oil	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	2%	2%	2%	2%	1%	1%	1%	1%	1%	1%	1%

Table S18. Breakdown of the India region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	13%	23%	28%	30%	34%	37%	43%	47%	50%	53%	57%
Hydro	10%	9%	8%	7%	6%	6%	5%	4%	3%	3%	3%	2%
Wind	3%	8%	13%	18%	20%	22%	24%	29%	34%	35%	36%	36%
Gas	2%	1%	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	74%	58%	43%	31%	24%	17%	12%	5%	1%	1%	1%	1%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	4%	6%	10%	14%	18%	20%	21%	19%	16%	12%	8%	4%
Oil	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	3%	3%	2%	2%	1%	1%	1%	0%	0%	0%	0%	0%
NetZero												
Solar	3%	17%	34%	42%	46%	46%	47%	50%	53%	55%	56%	58%
Hydro	10%	9%	7%	6%	5%	4%	3%	3%	2%	2%	2%	2%
Wind	3%	10%	20%	30%	34%	35%	35%	34%	34%	35%	37%	37%
Gas	2%	1%	1%	1%	1%	0%	0%	0%	0%	0%	0%	0%
Coal	74%	53%	26%	7%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	4%	6%	9%	12%	14%	13%	13%	12%	10%	8%	5%	2%
Oil	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	3%	3%	2%	1%	1%	1%	1%	1%	1%	1%	1%	1%

Table S19. Breakdown of the Japan region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	8%	11%	11%	12%	12%	12%	13%	14%	16%	17%	16%	18%
Hydro	12%	12%	13%	13%	12%	12%	13%	13%	13%	12%	12%	12%
Wind	1%	3%	6%	13%	20%	28%	35%	48%	58%	63%	65%	64%
Gas	27%	19%	13%	10%	6%	4%	2%	0%	0%	0%	0%	0%
Coal	28%	25%	24%	21%	20%	18%	16%	10%	5%	1%	0%	0%
Geothermal	1%	1%	1%	1%	1%	1%	1%	2%	2%	2%	2%	2%
Nuclear	7%	12%	16%	16%	14%	11%	8%	3%	1%	1%	1%	0%
Oil	9%	6%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	7%	11%	14%	14%	14%	14%	13%	11%	6%	4%	4%	5%
NetZero												
Solar	8%	15%	19%	20%	19%	18%	17%	16%	17%	19%	20%	19%
Hydro	12%	13%	13%	12%	11%	11%	11%	12%	11%	11%	11%	11%
Wind	1%	5%	18%	37%	46%	52%	57%	64%	67%	66%	65%	66%
Gas	27%	20%	13%	4%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	28%	16%	9%	1%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	2%
Nuclear	7%	13%	16%	15%	13%	9%	6%	2%	1%	1%	0%	0%

Richard Emmanuel Cabrera Jiménez

Oil	9%	6%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	7%	9%	9%	9%	8%	8%	7%	5%	2%	2%	3%	3%

Table S20. Breakdown of the Middle east region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	1%	4%	10%	15%	20%	24%	28%	34%	41%	48%	53%	55%
Hydro	5%	4%	3%	3%	2%	2%	2%	1%	1%	1%	1%	1%
Wind	1%	2%	5%	7%	7%	9%	11%	21%	35%	42%	45%	44%
Gas	81%	83%	79%	74%	70%	65%	59%	42%	23%	9%	1%	0%
Coal	3%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	1%	2%	1%	1%	1%	1%	1%	0%	0%	0%	0%	0%
Oil	9%	5%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
NetZero												
Solar	1%	12%	34%	51%	58%	55%	52%	49%	49%	53%	56%	57%
Hydro	5%	5%	4%	3%	2%	2%	2%	1%	1%	1%	0%	0%
Wind	1%	5%	14%	29%	38%	41%	44%	48%	48%	45%	43%	42%
Gas	81%	70%	45%	15%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	3%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	1%	2%	2%	2%	2%	2%	2%	1%	1%	0%	0%	0%
Oil	9%	6%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	0%	0%	0%	0%	0%	0%	0%	1%	1%	1%	1%	0%

Table S21. Breakdown of the Non-EU Europe countries region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	2%	6%	11%	16%	19%	22%	22%	21%	21%	24%	26%	27%
Hydro	51%	51%	48%	43%	41%	39%	36%	33%	30%	28%	25%	23%
Wind	6%	10%	13%	16%	20%	25%	30%	37%	40%	42%	45%	48%
Gas	13%	12%	11%	10%	8%	6%	4%	3%	2%	1%	0%	0%
Coal	20%	12%	8%	6%	3%	0%	0%	0%	0%	0%	0%	0%
Geothermal	3%	5%	5%	5%	5%	5%	5%	4%	4%	2%	1%	0%
Nuclear	4%	3%	3%	2%	2%	1%	1%	0%	0%	0%	0%	0%
Oil	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	1%	1%	1%	2%	3%	3%	2%	2%	2%	2%	2%
NetZero												
Solar	2%	11%	22%	27%	30%	30%	29%	27%	28%	30%	31%	31%
Hydro	51%	53%	43%	33%	26%	21%	18%	16%	14%	13%	12%	12%
Wind	6%	14%	23%	32%	39%	44%	48%	53%	55%	55%	55%	56%
Gas	13%	12%	4%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	20%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	3%	5%	5%	4%	3%	3%	2%	2%	2%	1%	1%	0%
Nuclear	4%	3%	2%	2%	1%	1%	0%	0%	0%	0%	0%	0%

Oil	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%	1%

Table S22. Breakdown of the Other Asian countries region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	10%	17%	22%	26%	29%	32%	35%	38%	41%	43%	43%
Hydro	16%	15%	13%	11%	9%	8%	7%	6%	5%	4%	4%	3%
Wind	1%	3%	9%	16%	22%	28%	34%	43%	50%	51%	51%	52%
Gas	32%	30%	28%	25%	21%	17%	13%	8%	3%	0%	0%	0%
Coal	32%	28%	22%	16%	12%	9%	7%	3%	0%	0%	0%	0%
Geothermal	3%	4%	3%	3%	2%	2%	2%	1%	1%	1%	1%	1%
Nuclear	9%	8%	7%	6%	5%	5%	4%	3%	2%	1%	1%	0%
Oil	4%	2%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	0%	1%	2%	2%	2%	2%	2%	1%	1%	0%	0%	1%
NetZero												
Solar	3%	15%	30%	39%	41%	41%	41%	41%	42%	43%	45%	48%
Hydro	16%	15%	13%	10%	8%	6%	5%	4%	3%	3%	2%	2%
Wind	1%	5%	18%	35%	43%	47%	49%	51%	52%	52%	51%	49%
Gas	32%	27%	20%	7%	0%	0%	0%	0%	0%	0%	0%	0%
Coal	32%	23%	7%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	3%	4%	3%	2%	2%	1%	1%	1%	1%	1%	1%	1%
Nuclear	9%	8%	7%	6%	5%	4%	3%	3%	2%	1%	1%	0%
Oil	4%	2%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	0%	1%	2%	1%	1%	1%	1%	1%	1%	1%	1%	1%

Table S23. Breakdown of the Reforming economies region electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	1%	2%	4%	6%	8%	10%	13%	18%	23%	29%	33%	37%
Hydro	17%	16%	14%	12%	10%	10%	9%	9%	8%	8%	8%	7%
Wind	1%	1%	1%	1%	1%	1%	1%	5%	14%	26%	37%	43%
Gas	33%	36%	43%	50%	56%	60%	62%	61%	48%	32%	18%	8%
Coal	26%	24%	19%	15%	11%	8%	5%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	20%	18%	15%	11%	9%	7%	5%	3%	1%	0%	0%	0%
Oil	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	2%	4%	4%	4%	4%	4%	4%	5%	5%	4%	4%
NetZero												
Solar	1%	7%	19%	28%	33%	35%	35%	36%	36%	38%	39%	41%
Hydro	17%	19%	16%	13%	12%	11%	10%	9%	8%	8%	8%	7%
Wind	1%	4%	14%	28%	38%	42%	44%	47%	50%	52%	51%	51%
Gas	33%	39%	30%	14%	3%	0%	0%	0%	0%	0%	0%	0%
Coal	26%	5%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Nuclear	20%	23%	18%	14%	11%	9%	7%	5%	3%	1%	0%	0%

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Oil	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	1%	3%	4%	4%	4%	4%	4%	3%	2%	2%	1%	1%

Table S24. Breakdown of Global electricity mix.

Current policies	2020	2025	2030	2035	2040	2045	2050	2060	2070	2080	2090	2100
Solar	3%	8%	15%	19%	22%	25%	27%	29%	31%	34%	36%	37%
Hydro	24%	25%	22%	20%	18%	17%	16%	15%	14%	13%	12%	11%
Wind	5%	9%	13%	17%	21%	25%	29%	36%	42%	45%	47%	47%
Gas	21%	20%	19%	19%	17%	16%	14%	11%	7%	4%	2%	1%
Coal	31%	23%	17%	12%	10%	7%	5%	2%	1%	1%	0%	0%
Geothermal	1%	2%	2%	2%	1%	1%	1%	1%	1%	1%	0%	0%
Nuclear	10%	9%	8%	8%	7%	6%	5%	4%	3%	2%	1%	1%
Oil	3%	2%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	3%	3%	3%	3%	3%	3%	2%	2%	2%	2%	2%
NetZero												
Solar	3%	14%	26%	33%	36%	36%	36%	35%	36%	38%	40%	41%
Hydro	24%	26%	22%	18%	15%	14%	13%	12%	11%	10%	9%	9%
Wind	5%	12%	22%	32%	38%	42%	44%	47%	49%	49%	49%	49%
Gas	21%	19%	11%	4%	1%	0%	0%	0%	0%	0%	0%	0%
Coal	31%	12%	4%	1%	0%	0%	0%	0%	0%	0%	0%	0%
Geothermal	1%	2%	2%	1%	1%	1%	1%	1%	1%	1%	0%	0%
Nuclear	10%	10%	9%	7%	6%	5%	4%	3%	2%	2%	1%	0%
Oil	3%	2%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Biomass	2%	3%	3%	3%	3%	2%	2%	2%	1%	1%	1%	1%

3. Additional result information

3.1. Global warming potential emissions

Table S25. GHG emissions for European Union and UK region scenarios.

European Union and UK region CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,08	0,06	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	4,3E+12	4,3E+12	4,2E+12	4,2E+12	3,9E+12	3,8E+12	3,6E+12	3,6E+12	3,6E+12	3,6E+12	3,6E+12	3,7E+12	3,7E+12
BET annual tkm transported	0,0E+00	1,6E+11	6,6E+11	1,5E+12	2,1E+12	2,3E+12	2,5E+12	2,6E+12	2,7E+12	2,9E+12	3,0E+12	3,1E+12	3,2E+12
ICT annual tkm transported	4,3E+12	4,1E+12	3,6E+12	2,7E+12	1,9E+12	1,4E+12	1,2E+12	9,5E+11	8,4E+11	7,2E+11	6,5E+11	6,0E+11	5,3E+11
Model IAM [kg CO ₂ /year]	414,4	359,7	313,7	282,7	234,0	194,9	168,0	159,3	153,0	148,9	148,1	148,4	146,1
Electric based scenario [kg CO ₂ /year]	466,3	337,0	266,1	220,9	180,5	149,8	130,4	129,2	127,5	128,1	129,9	132,2	132,7
Biofuel based scenario [kg CO ₂ /year]	289,5	257,2	234,0	230,1	214,7	200,4	187,9	187,4	186,2	187,0	188,8	191,7	192,2
Diesel based scenario [kg CO ₂ /year]	421,9	368,4	330,3	324,3	302,0	280,7	262,2	261,6	259,9	261,0	263,5	267,5	268,2
European Union and UK region NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,07	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	4,3E+12	4,2E+12	3,8E+12	3,3E+12	2,8E+12	2,5E+12	2,3E+12	2,2E+12	2,2E+12	2,2E+12	2,2E+12	2,2E+12	2,3E+12
BET annual tkm transported	0,0E+00	2,2E+11	8,1E+11	1,5E+12	1,8E+12	1,8E+12	1,7E+12	1,8E+12	1,8E+12	1,8E+12	1,8E+12	1,9E+12	2,0E+12
ICT annual tkm transported	4,3E+12	3,9E+12	3,0E+12	1,9E+12	1,0E+12	6,7E+11	5,5E+11	4,7E+11	4,2E+11	3,7E+11	3,4E+11	3,1E+11	2,7E+11
Model IAM [kg CO ₂ /year]	413,1	347,5	270,1	205,3	144,9	114,4	97,6	92,8	89,4	86,2	85,5	86,6	86,2
Electric based scenario [Mt CO ₂ /year]	464,9	284,6	200,6	154,5	116,7	95,8	82,2	80,5	78,9	77,9	78,3	80,1	80,9
Biofuel based scenario [Mt CO ₂ /year]	288,6	250,7	210,6	183,0	151,1	132,0	119,4	117,2	115,2	113,6	113,8	116,4	117,6
Diesel based scenario [Mt CO ₂ /year]	420,6	359,1	297,2	257,9	212,5	184,9	166,7	163,6	160,7	158,6	158,9	162,5	164,2

Table S26.GHG emissions for Latin America region scenarios.

Latin America region CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,10	0,07	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	3,2E+12	3,7E+12	4,2E+12	4,9E+12	5,3E+12	5,5E+12	5,6E+12	5,6E+12	5,6E+12	5,4E+12	5,6E+12	5,6E+12	5,9E+12
BET annual tkm transported	0,0E+00	3,3E+10	1,5E+11	4,7E+11	9,3E+11	1,5E+12	2,1E+12	2,7E+12	3,1E+12	3,5E+12	3,9E+12	3,9E+12	4,1E+12
ICT annual tkm transported	3,2E+12	3,6E+12	4,1E+12	4,4E+12	4,3E+12	4,0E+12	3,5E+12	2,9E+12	2,4E+12	1,9E+12	1,7E+12	1,7E+12	1,8E+12
Model IAM [kg CO ₂ /year]	315,2	312,6	326,8	362,4	369,9	355,7	327,6	304,1	283,7	252,3	247,5	238,6	245,2
Electric based scenario [kg CO ₂ /year]	326,1	272,6	252,5	245,8	235,1	222,5	207,2	205,5	202,9	195,2	202,0	201,9	211,4
Biofuel based scenario [kg CO ₂ /year]	218,4	220,4	235,1	267,6	285,6	292,4	290,3	291,0	290,6	282,2	292,0	292,1	305,5
Diesel based scenario [kg CO ₂ /year]	318,3	315,8	331,9	377,1	401,8	409,7	405,2	406,2	405,6	393,8	407,5	407,7	426,3
Latin America region NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,10	0,07	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	3,2E+12	3,4E+12	3,7E+12	4,1E+12	4,1E+12	4,2E+12	4,3E+12	4,4E+12	4,6E+12	4,5E+12	4,7E+12	4,8E+12	5,0E+12
BET annual tkm transported	0,0E+00	9,1E+10	5,2E+11	1,5E+12	2,5E+12	3,2E+12	3,4E+12	3,6E+12	3,8E+12	3,9E+12	4,3E+12	4,4E+12	4,7E+12
ICT annual tkm transported	3,2E+12	3,3E+12	3,2E+12	2,5E+12	1,5E+12	1,0E+12	8,8E+11	8,2E+11	7,5E+11	5,8E+11	4,8E+11	3,7E+11	3,0E+11
Model IAM [kg CO ₂ /year]	312,1	287,9	273,6	254,5	207,4	182,9	174,8	177,7	180,9	173,8	181,1	181,3	188,4
Electric based scenario [Mt CO ₂ /year]	322,9	226,9	202,6	189,6	170,3	160,9	154,1	158,2	163,6	160,9	170,4	172,9	181,7
Biofuel based scenario [Mt CO ₂ /year]	216,3	204,3	207,9	223,5	220,3	221,4	223,4	229,4	237,3	233,4	246,7	250,1	262,3
Diesel based scenario [Mt CO ₂ /year]	315,2	292,7	293,4	314,9	310,0	310,2	311,7	320,2	331,2	325,7	344,3	349,1	366,1

Table S27.GHG emissions for China region scenarios.

China region CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,16	0,12	0,10	0,08	0,07	0,06	0,05	0,05	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	7,5E+12	7,2E+12	7,1E+12	7,3E+12	6,9E+12	6,5E+12	6,3E+12	6,2E+12	6,2E+12	6,2E+12	6,2E+12	6,1E+12	6,2E+12
BET annual tkm transported	1,8E+11	3,9E+11	8,0E+11	1,7E+12	2,5E+12	3,2E+12	3,6E+12	4,1E+12	4,4E+12	4,8E+12	5,0E+12	5,1E+12	5,4E+12
ICT annual tkm transported	7,3E+12	6,8E+12	6,3E+12	5,6E+12	4,4E+12	3,3E+12	2,7E+12	2,1E+12	1,8E+12	1,4E+12	1,2E+12	9,9E+11	8,5E+11
Model IAM [kg CO ₂ /year]	744,3	634,7	569,6	568,4	509,0	444,0	383,0	363,6	324,8	295,3	273,4	256,6	252,6
Electric based scenario [kg CO ₂ /year]	1172,6	846,5	682,3	590,6	481,2	401,7	331,9	323,5	278,3	254,4	238,3	226,2	228,0
Biofuel based scenario [kg CO ₂ /year]	503,8	434,7	393,4	399,0	373,5	347,4	328,2	323,5	325,2	325,1	322,1	318,8	325,1
Diesel based scenario [kg CO ₂ /year]	734,3	622,8	555,3	562,2	525,5	486,7	458,0	451,4	453,9	453,7	449,5	445,0	453,7
China region NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,16	0,09	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	7,6E+12	7,6E+12	7,7E+12	7,9E+12	7,3E+12	6,7E+12	6,1E+12	5,8E+12	5,7E+12	5,6E+12	5,6E+12	5,6E+12	5,7E+12
BET annual tkm transported	5,8E+11	1,3E+12	2,3E+12	3,9E+12	5,0E+12	5,6E+12	5,7E+12	5,6E+12	5,5E+12	5,7E+12	5,7E+12	5,9E+12	6,1E+12
ICT annual tkm transported	7,2E+12	6,6E+12	5,7E+12	4,5E+12	2,8E+12	1,6E+12	1,0E+12	8,0E+11	7,3E+11	5,9E+11	4,7E+11	4,0E+11	3,2E+11
Model IAM [kg CO ₂ /year]	792,2	683,0	586,2	532,5	395,9	277,4	234,6	223,7	219,6	220,4	220,6	226,0	231,3
Electric based scenario [Mt CO ₂ /year]	1188,3	683,0	460,0	385,0	316,1	259,4	222,3	210,4	205,2	202,6	201,9	205,3	207,7
Biofuel based scenario [Mt CO ₂ /year]	510,6	456,5	426,7	435,9	399,4	354,1	320,0	303,6	297,1	293,8	291,1	294,5	298,3
Diesel based scenario [Mt CO ₂ /year]	744,2	654,0	602,3	614,3	561,9	496,1	446,6	423,8	414,6	410,1	406,2	411,0	416,2

Table S28. GHG emissions for USA region scenarios.

USA region CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,13	0,10	0,08	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	4,0E+12	3,9E+12	3,8E+12	3,6E+12	3,4E+12	3,1E+12	2,9E+12	2,8E+12	2,6E+12	2,6E+12	2,6E+12	2,6E+12	2,6E+12
BET annual tkm transported	5,8E+10	1,4E+11	2,8E+11	5,3E+11	8,1E+11	1,0E+12	1,2E+12	1,3E+12	1,5E+12	1,7E+12	1,8E+12	2,0E+12	2,1E+12
ICT annual tkm transported	4,0E+12	3,7E+12	3,5E+12	3,1E+12	2,6E+12	2,1E+12	1,7E+12	1,4E+12	1,2E+12	9,2E+11	7,4E+11	6,4E+11	5,7E+11
Model IAM [kg CO ₂ /year]	393,2	336,7	296,5	273,6	237,3	198,8	168,5	154,0	140,3	126,8	118,8	115,7	113,7
Electric based scenario [kg CO ₂ /year]	536,0	405,4	301,0	226,3	176,8	137,7	113,1	106,6	99,8	94,4	93,5	94,9	96,9
Biofuel based scenario [kg CO ₂ /year]	269,6	234,3	210,3	200,4	182,9	164,0	148,6	143,5	137,7	134,7	133,3	135,1	137,9
Diesel based scenario [kg CO ₂ /year]	393,0	335,7	296,8	282,4	257,4	229,7	207,4	200,3	192,2	188,1	186,0	188,6	192,4
USA region NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,13	0,08	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	4,0E+12	4,0E+12	3,9E+12	3,7E+12	3,1E+12	2,6E+12	2,3E+12	2,3E+12	2,2E+12	2,1E+12	2,1E+12	2,2E+12	2,2E+12
BET annual tkm transported	1,8E+11	4,1E+11	7,5E+11	1,1E+12	1,4E+12	1,5E+12	1,4E+12	1,5E+12	1,5E+12	1,7E+12	1,8E+12	1,9E+12	2,0E+12
ICT annual tkm transported	3,9E+12	3,6E+12	3,2E+12	2,5E+12	1,7E+12	1,2E+12	9,5E+11	8,0E+11	6,6E+11	4,6E+11	3,4E+11	2,8E+11	2,4E+11
Model IAM [kg CO ₂ /year]	401,4	340,8	281,9	230,5	165,6	127,6	105,8	99,2	93,4	87,6	85,6	86,4	87,4
Electric based scenario [Mt CO ₂ /year]	541,1	339,8	233,8	176,5	132,2	104,2	85,6	82,5	79,8	77,9	78,3	80,4	82,3
Biofuel based scenario [Mt CO ₂ /year]	272,2	241,4	217,4	202,0	167,9	140,8	121,9	117,6	113,7	111,1	111,2	114,1	116,8
Diesel based scenario [Mt CO ₂ /year]	396,7	345,8	306,9	284,6	236,2	197,3	170,1	164,1	158,6	155,1	155,2	159,2	163,0

Table S29. GHG emissions for Canada, Australia and New Zealand region scenarios.

Canada, Australia and New Zealand CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,08	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	9,3E+11	9,2E+11	8,7E+11	8,3E+11	7,7E+11	7,1E+11	6,8E+11	6,6E+11	6,6E+11	6,5E+11	6,6E+11	6,6E+11	6,7E+11
BET annual tkm transported	0,0E+00	1,3E+10	5,4E+10	1,4E+11	2,2E+11	2,7E+11	3,2E+11	3,6E+11	4,0E+11	4,3E+11	4,6E+11	4,9E+11	5,2E+11
ICT annual tkm transported	9,3E+11	9,1E+11	8,2E+11	7,0E+11	5,5E+11	4,4E+11	3,6E+11	3,0E+11	2,6E+11	2,2E+11	1,9E+11	1,7E+11	1,5E+11
Model IAM [kg CO ₂ /year]	91,3	79,2	67,0	60,7	51,5	43,1	37,2	34,2	32,4	29,9	28,0	26,2	26,4
Electric based scenario [kg CO ₂ /year]	100,1	69,0	52,4	42,2	33,9	27,2	23,9	23,3	23,1	22,8	23,1	23,3	23,9
Biofuel based scenario [kg CO ₂ /year]	62,7	55,5	48,3	45,8	41,7	38,0	35,2	34,5	34,2	33,7	34,2	34,5	35,1
Diesel based scenario [kg CO ₂ /year]	91,4	79,5	68,1	64,6	58,6	53,2	49,2	48,1	47,7	47,1	47,7	48,1	49,0
Canada, Australia and New Zealand NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,06	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	9,2E+11	9,0E+11	8,5E+11	8,1E+11	6,9E+11	6,1E+11	5,5E+11	5,4E+11	5,4E+11	5,4E+11	5,4E+11	5,5E+11	5,7E+11
BET annual tkm transported	0,0E+00	3,2E+10	1,3E+11	3,1E+11	4,0E+11	4,3E+11	4,1E+11	4,2E+11	4,3E+11	4,4E+11	4,7E+11	4,8E+11	5,0E+11
ICT annual tkm transported	9,2E+11	8,7E+11	7,1E+11	5,0E+11	2,8E+11	1,8E+11	1,5E+11	1,3E+11	1,2E+11	9,1E+10	7,7E+10	6,8E+10	6,4E+10
Model IAM [kg CO ₂ /year]	90,0	77,1	62,7	51,3	35,6	27,1	23,2	22,4	22,0	21,0	21,0	21,1	21,7
Electric based scenario [Mt CO ₂ /year]	98,7	57,8	44,0	36,9	28,5	23,1	19,6	19,3	19,2	18,9	19,2	19,5	20,3
Biofuel based scenario [Mt CO ₂ /year]	61,8	54,4	47,1	44,3	37,4	32,2	28,9	28,4	28,4	27,9	28,3	28,6	29,6
Diesel based scenario [Mt CO ₂ /year]	90,1	77,9	66,4	62,5	52,6	45,1	40,4	39,6	39,6	39,0	39,5	40,0	41,4

Table S30. GHG emissions for Reforming economies countries region scenarios.

Reforming economies CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,13	0,11	0,10	0,09	0,08	0,07	0,07	0,07	0,06	0,06	0,05	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	6,4E+11	7,2E+11	7,4E+11	7,3E+11	6,9E+11	6,3E+11	6,0E+11	6,1E+11	6,1E+11	6,1E+11	6,4E+11	6,3E+11	6,5E+11
BET annual tkm transported	0,0E+00	1,1E+10	4,5E+10	1,1E+11	1,7E+11	2,2E+11	2,4E+11	2,9E+11	3,1E+11	3,5E+11	4,0E+11	4,3E+11	4,7E+11
ICT annual tkm transported	6,4E+11	7,1E+11	7,0E+11	6,2E+11	5,1E+11	4,2E+11	3,6E+11	3,2E+11	3,0E+11	2,6E+11	2,4E+11	2,0E+11	1,8E+11
Model IAM [kg CO ₂ /year]	62,2	62,5	58,9	57,7	53,2	47,1	42,8	42,6	41,7	39,5	37,2	29,6	28,3
Electric based scenario [kg CO ₂ /year]	83,9	79,3	71,7	63,9	55,3	47,0	41,3	40,4	39,1	35,8	32,4	28,0	26,1
Biofuel based scenario [kg CO ₂ /year]	42,7	43,4	41,1	40,1	37,3	33,7	31,5	31,9	31,9	32,0	33,2	32,8	33,9
Diesel based scenario [kg CO ₂ /year]	62,2	62,2	58,0	56,5	52,5	47,2	43,9	44,5	44,5	44,6	46,3	45,8	47,3
Reforming economies NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,13	0,09	0,07	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	6,4E+11	7,1E+11	6,9E+11	6,6E+11	5,9E+11	5,3E+11	4,9E+11	4,9E+11	4,9E+11	4,9E+11	5,1E+11	5,1E+11	5,3E+11
BET annual tkm transported	0,0E+00	3,3E+10	1,2E+11	2,7E+11	3,5E+11	3,8E+11	3,7E+11	3,8E+11	3,9E+11	4,1E+11	4,4E+11	4,5E+11	4,8E+11
ICT annual tkm transported	6,4E+11	6,8E+11	5,7E+11	3,9E+11	2,4E+11	1,5E+11	1,2E+11	1,1E+11	9,8E+10	8,0E+10	7,1E+10	6,1E+10	5,4E+10
Model IAM [kg CO ₂ /year]	63,1	61,8	52,9	44,6	32,3	24,4	21,0	20,3	19,6	18,9	19,5	19,6	20,2
Electric based scenario [Mt CO ₂ /year]	85,1	65,1	48,3	35,8	25,5	20,5	17,8	17,7	17,6	17,6	18,4	18,6	19,4
Biofuel based scenario [Mt CO ₂ /year]	43,3	43,1	38,3	36,3	31,9	28,1	25,7	25,5	25,3	25,3	26,4	26,7	27,8
Diesel based scenario [Mt CO ₂ /year]	63,1	61,7	54,1	51,2	44,9	39,4	35,8	35,6	35,3	35,4	36,9	37,3	38,8

Table S31. GHG emissions for Other Asian countries region scenarios.

Other Asian countries CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,11	0,09	0,08	0,06	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,7E+12	3,7E+12	4,6E+12	5,4E+12	6,0E+12	6,6E+12	6,8E+12	7,2E+12	7,6E+12	8,1E+12	8,8E+12	9,9E+12	1,1E+13
BET annual tkm transported	0,0E+00	5,1E+10	2,5E+11	6,8E+11	1,2E+12	1,8E+12	2,3E+12	2,9E+12	3,4E+12	4,3E+12	5,1E+12	6,2E+12	7,3E+12
ICT annual tkm transported	2,7E+12	3,6E+12	4,3E+12	4,7E+12	4,8E+12	4,8E+12	4,5E+12	4,3E+12	4,2E+12	3,8E+12	3,7E+12	3,7E+12	3,7E+12
Model IAM [kg CO ₂ /year]	268,9	317,1	360,7	414,2	443,3	454,2	436,8	439,2	443,0	437,6	453,4	492,7	531,6
Electric based scenario [kg CO ₂ /year]	390,1	414,2	418,2	404,0	383,8	361,3	325,2	321,6	317,6	307,8	324,9	363,4	404,9
Biofuel based scenario [kg CO ₂ /year]	184,5	220,4	253,3	295,0	325,9	349,6	355,3	374,6	396,2	424,7	459,6	515,6	573,3
Diesel based scenario [kg CO ₂ /year]	269,0	315,8	357,5	415,7	458,5	489,9	495,8	522,8	553,0	592,7	641,4	719,6	800,0
Other Asian countries NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,11	0,07	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,7E+12	3,5E+12	4,2E+12	4,9E+12	5,3E+12	5,6E+12	5,5E+12	6,0E+12	6,4E+12	7,0E+12	7,7E+12	9,0E+12	9,8E+12
BET annual tkm transported	0,0E+00	6,8E+10	3,9E+11	1,3E+12	2,4E+12	3,4E+12	4,1E+12	4,8E+12	5,1E+12	5,7E+12	6,5E+12	7,8E+12	8,6E+12
ICT annual tkm transported	2,7E+12	3,4E+12	3,8E+12	3,6E+12	2,9E+12	2,1E+12	1,5E+12	1,2E+12	1,3E+12	1,3E+12	1,2E+12	1,2E+12	1,2E+12
Model IAM [kg CO ₂ /year]	265,6	301,0	325,7	342,6	324,5	288,5	249,2	259,9	274,2	296,6	317,9	364,0	393,0
Electric based scenario [Mt CO ₂ /year]	385,2	369,0	305,8	247,0	226,7	217,5	201,8	219,9	233,5	256,5	281,3	329,2	360,5
Biofuel based scenario [Mt CO ₂ /year]	182,2	209,2	232,5	267,3	288,5	295,6	288,9	314,7	334,0	366,7	401,3	468,2	510,9
Diesel based scenario [Mt CO ₂ /year]	265,6	299,8	328,2	376,6	405,8	414,2	403,2	439,2	466,1	511,7	560,1	653,5	713,0

Table S32. GHG emissions for non-EU Europe countries region scenarios.

non-EU Europe countries CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,08	0,07	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	5,7E+11	6,2E+11	6,6E+11	6,7E+11	6,5E+11	6,3E+11	6,1E+11	6,2E+11	6,2E+11	6,2E+11	6,2E+11	6,4E+11	6,4E+11
BET annual tkm transported	0,0E+00	1,0E+10	4,8E+10	1,3E+11	2,2E+11	2,7E+11	3,0E+11	3,4E+11	3,6E+11	4,0E+11	4,3E+11	4,6E+11	4,8E+11
ICT annual tkm transported	5,7E+11	6,1E+11	6,1E+11	5,3E+11	4,3E+11	3,6E+11	3,2E+11	2,8E+11	2,6E+11	2,2E+11	1,9E+11	1,8E+11	1,6E+11
Model IAM [kg CO ₂ /year]	55,6	53,3	50,9	48,7	43,2	37,9	34,1	32,9	31,7	29,9	27,7	27,1	26,2
Electric based scenario [kg CO ₂ /year]	63,1	51,1	43,7	37,7	31,2	25,9	23,0	23,0	22,7	22,4	22,5	22,7	22,7
Biofuel based scenario [kg CO ₂ /year]	38,1	37,3	36,5	36,6	35,1	33,6	32,0	32,3	32,2	32,1	32,4	33,1	33,2
Diesel based scenario [kg CO ₂ /year]	55,6	53,4	51,5	51,6	49,4	47,1	44,7	45,0	44,9	44,8	45,2	46,2	46,3
non-EU Europe countries NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,11	0,07	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	5,8E+11	6,6E+11	6,9E+11	6,9E+11	6,4E+11	6,0E+11	5,6E+11	5,7E+11	5,7E+11	5,7E+11	5,8E+11	6,0E+11	5,9E+11
BET annual tkm transported	0,0E+00	2,8E+10	1,2E+11	2,9E+11	3,9E+11	4,2E+11	4,2E+11	4,3E+11	4,5E+11	4,8E+11	5,0E+11	5,2E+11	5,2E+11
ICT annual tkm transported	5,8E+11	6,4E+11	5,7E+11	4,0E+11	2,5E+11	1,8E+11	1,5E+11	1,3E+11	1,2E+11	9,5E+10	8,3E+10	7,7E+10	7,2E+10
Model IAM [kg CO ₂ /year]	56,8	56,6	50,7	43,8	34,4	27,0	23,7	23,4	23,0	22,0	22,3	22,6	22,5
Electric based scenario [Mt CO ₂ /year]	64,5	46,0	37,3	32,0	26,9	23,2	20,2	20,3	20,4	20,5	20,9	21,4	21,3
Biofuel based scenario [Mt CO ₂ /year]	39,0	40,0	38,2	38,0	35,0	32,0	29,5	29,5	29,8	29,8	30,4	31,0	31,0
Diesel based scenario [Mt CO ₂ /year]	56,8	57,4	53,9	53,5	49,3	44,8	41,1	41,2	41,5	41,6	42,4	43,3	43,3

Table S33. GHG emissions for sub-Saharan Africa region scenarios.

sub-Saharan Africa region CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,10	0,08	0,07	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	9,8E+11	1,2E+12	1,8E+12	2,6E+12	3,7E+12	5,3E+12	7,4E+12	1,0E+13	1,4E+13	1,9E+13	2,2E+13	2,4E+13	2,6E+13
BET annual tkm transported	0,0E+00	3,1E+09	2,2E+10	1,1E+11	3,1E+11	7,0E+11	1,4E+12	2,6E+12	4,2E+12	6,8E+12	8,8E+12	1,1E+13	1,3E+13
ICT annual tkm transported	9,8E+11	1,2E+12	1,8E+12	2,5E+12	3,4E+12	4,6E+12	6,0E+12	7,8E+12	1,0E+13	1,2E+13	1,3E+13	1,3E+13	1,3E+13
Model IAM [kg CO ₂ /year]	96,0	106,2	139,3	200,8	279,0	378,4	494,7	677,7	913,3	1123,8	1269,5	1352,0	1427,3
Electric based scenario [kg CO ₂ /year]	135,9	125,0	141,2	172,7	214,0	262,1	319,5	435,8	582,9	721,8	827,4	903,5	985,7
Biofuel based scenario [kg CO ₂ /year]	65,9	74,1	98,7	143,4	202,6	282,7	383,3	542,0	752,3	969,6	1127,0	1228,5	1329,4
Diesel based scenario [kg CO ₂ /year]	96,0	106,2	139,3	202,0	285,0	396,0	534,9	756,4	1050,0	1353,2	1572,9	1714,6	1855,4
sub-Saharan Africa region NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,09	0,06	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	9,7E+11	1,2E+12	1,7E+12	2,3E+12	2,9E+12	3,7E+12	4,7E+12	6,9E+12	9,8E+12	1,3E+13	1,6E+13	1,8E+13	2,0E+13
BET annual tkm transported	0,0E+00	3,5E+09	3,0E+10	1,8E+11	6,0E+11	1,4E+12	2,5E+12	4,3E+12	6,4E+12	9,3E+12	1,2E+13	1,4E+13	1,6E+13
ICT annual tkm transported	9,7E+11	1,2E+12	1,7E+12	2,1E+12	2,3E+12	2,3E+12	2,2E+12	2,6E+12	3,3E+12	3,8E+12	4,1E+12	4,1E+12	4,1E+12
Model IAM [kg CO ₂ /year]	95,3	102,0	131,1	167,2	194,4	213,1	229,4	319,5	441,0	568,4	664,3	744,0	825,5
Electric based scenario [Mt CO ₂ /year]	134,9	107,4	107,2	108,5	123,8	142,5	170,5	251,7	357,4	482,1	581,2	662,5	742,4
Biofuel based scenario [Mt CO ₂ /year]	65,4	71,4	93,8	126,0	159,3	194,5	244,2	359,9	509,9	686,9	825,8	937,6	1045,9
Diesel based scenario [Mt CO ₂ /year]	95,3	102,3	132,5	177,6	224,1	272,5	340,9	502,2	711,7	958,7	1152,5	1308,5	1459,7

Table S34. GHG emissions for India region scenarios.

India CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,17	0,13	0,10	0,08	0,06	0,05	0,05	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,3E+12	3,4E+12	4,5E+12	5,8E+12	6,8E+12	7,5E+12	7,9E+12	7,9E+12	8,0E+12	8,2E+12	8,4E+12	8,4E+12	8,4E+12
BET annual tkm transported	0,0E+00	1,2E+10	8,3E+10	3,0E+11	6,4E+11	1,1E+12	1,6E+12	2,1E+12	2,4E+12	2,7E+12	2,9E+12	3,1E+12	3,5E+12
ICT annual tkm transported	2,3E+12	3,3E+12	4,5E+12	5,5E+12	6,1E+12	6,4E+12	6,2E+12	5,8E+12	5,6E+12	5,5E+12	5,5E+12	5,3E+12	4,9E+12
Model IAM [kg CO ₂ /year]	228,7	290,7	357,5	448,1	510,4	534,3	527,3	509,1	501,0	501,1	507,5	500,0	488,6
Electric based scenario [kg CO ₂ /year]	391,2	426,4	444,0	443,8	433,5	397,0	359,4	337,7	319,3	305,9	312,7	313,3	317,6
Biofuel based scenario [kg CO ₂ /year]	157,0	202,6	252,1	318,2	368,5	398,3	409,5	410,0	414,8	428,7	437,5	436,3	439,2
Diesel based scenario [kg CO ₂ /year]	228,8	290,2	355,9	448,4	518,5	558,0	571,5	572,2	578,9	598,3	610,6	608,9	613,0
India NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,17	0,12	0,08	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,3E+12	3,2E+12	3,9E+12	4,4E+12	4,8E+12	4,9E+12	4,7E+12	5,0E+12	5,1E+12	5,4E+12	5,4E+12	5,8E+12	5,9E+12
BET annual tkm transported	0,0E+00	1,6E+10	1,2E+11	5,5E+11	1,4E+12	2,4E+12	3,2E+12	3,8E+12	3,8E+12	4,1E+12	4,3E+12	4,6E+12	4,6E+12
ICT annual tkm transported	2,3E+12	3,2E+12	3,7E+12	3,9E+12	3,4E+12	2,5E+12	1,5E+12	1,2E+12	1,3E+12	1,3E+12	1,2E+12	1,2E+12	1,2E+12
Model IAM [kg CO ₂ /year]	222,5	276,2	303,3	328,8	318,9	273,4	214,8	219,1	225,9	236,9	228,3	245,4	250,4
Electric based scenario [Mt CO ₂ /year]	380,5	387,6	313,9	239,6	205,4	192,2	172,4	184,8	189,1	200,3	201,4	215,7	218,5
Biofuel based scenario [Mt CO ₂ /year]	152,7	192,4	214,7	242,7	259,7	258,7	244,3	261,8	267,8	282,6	283,2	302,5	305,4

Table S35. GHG emissions for Japan region scenarios.

Japan CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,11	0,09	0,07	0,06	0,06	0,05	0,05	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	1,1E+12	1,1E+12	1,1E+12	9,8E+11	8,5E+11	7,6E+11	7,0E+11	6,8E+11	6,6E+11	6,5E+11	6,5E+11	6,5E+11	6,6E+11
BET annual tkm transported	0,0E+00	2,2E+10	9,2E+10	2,0E+11	2,9E+11	3,3E+11	3,5E+11	3,9E+11	4,2E+11	4,6E+11	4,9E+11	5,1E+11	5,3E+11
ICT annual tkm transported	1,1E+12	1,1E+12	9,6E+11	7,8E+11	5,6E+11	4,3E+11	3,5E+11	2,9E+11	2,5E+11	2,0E+11	1,7E+11	1,4E+11	1,2E+11
Model IAM [kg CO ₂ /year]	107,5	94,0	83,2	75,0	61,2	50,2	42,4	38,7	35,7	31,3	28,5	27,3	26,8
Electric based scenario [kg CO ₂ /year]	153,8	116,1	90,1	70,6	53,4	42,0	34,3	31,5	29,0	25,8	23,7	23,1	23,3
Biofuel based scenario [kg CO ₂ /year]	73,7	65,3	58,5	54,0	46,3	40,4	36,6	35,5	34,6	33,9	34,1	34,0	34,3
Diesel based scenario [kg CO ₂ /year]	107,5	93,5	82,5	76,1	65,2	56,7	51,0	49,5	48,3	47,4	47,5	47,4	47,9
Japan NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,14	0,10	0,07	0,05	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	1,1E+12	1,2E+12	1,1E+12	1,0E+12	8,8E+11	7,4E+11	6,3E+11	6,0E+11	5,9E+11	6,0E+11	6,0E+11	6,0E+11	6,2E+11
BET annual tkm transported	0,0E+00	4,2E+10	1,6E+11	3,4E+11	4,6E+11	4,8E+11	4,4E+11	4,4E+11	4,5E+11	4,8E+11	5,1E+11	5,2E+11	5,4E+11
ICT annual tkm transported	1,1E+12	1,1E+12	9,9E+11	7,0E+11	4,2E+11	2,6E+11	1,9E+11	1,6E+11	1,4E+11	1,1E+11	9,5E+10	8,1E+10	7,2E+10
Model IAM [kg CO ₂ /year]	112,4	102,8	87,5	69,5	48,8	34,4	26,9	24,9	24,4	23,8	23,5	23,2	23,4
Electric based scenario [Mt CO ₂ /year]	160,8	118,5	81,1	51,0	36,4	28,2	22,4	21,1	21,0	21,0	21,2	21,3	21,8
Biofuel based scenario [Mt CO ₂ /year]	77,1	71,9	63,8	56,8	47,7	39,4	33,0	31,1	31,0	31,1	31,3	31,3	32,1
Diesel based scenario [Mt CO ₂ /year]	112,4	103,0	90,0	80,1	67,2	55,1	46,0	43,4	43,2	43,3	43,6	43,7	44,8

Table S36. GHG emissions for Middle east region scenarios.

Middle east CP	2020	2025	2030	2035	2040	2045	2050	2055	2060	2070	2080	2090	2100
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,15	0,12	0,10	0,09	0,08	0,07	0,06	0,06	0,06	0,05	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,2E+12	2,7E+12	3,1E+12	3,7E+12	4,2E+12	4,7E+12	5,1E+12	5,7E+12	6,1E+12	6,3E+12	6,6E+12	6,8E+12	7,2E+12
BET annual tkm transported	0,0E+00	1,0E+10	5,4E+10	2,0E+11	4,3E+11	6,8E+11	9,8E+11	1,4E+12	1,7E+12	2,1E+12	2,5E+12	3,0E+12	3,5E+12
ICT annual tkm transported	2,2E+12	2,7E+12	3,1E+12	3,5E+12	3,8E+12	4,0E+12	4,2E+12	4,3E+12	4,4E+12	4,3E+12	4,1E+12	3,8E+12	3,7E+12
Model IAM [kg CO ₂ /year]	211,5	236,5	246,9	288,9	325,2	345,8	365,1	395,1	415,2	405,0	395,0	390,2	398,1
Electric based scenario [kg CO ₂ /year]	315,6	324,9	314,6	323,3	332,0	329,9	325,9	339,1	342,0	299,9	268,6	256,4	269,2
Biofuel based scenario [kg CO ₂ /year]	145,1	164,8	174,1	203,7	230,8	248,9	268,4	296,1	317,9	329,6	341,9	354,9	376,7
Diesel based scenario [kg CO ₂ /year]	211,5	236,1	245,8	287,1	324,7	348,8	374,5	413,2	443,6	460,0	477,1	495,2	525,7
Middle east NZ													
ICT diesel emissions [kg CO ₂ /tkm]	0,10	0,09	0,08	0,08	0,08	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07
BET emissions [kg CO ₂ /tkm]	0,15	0,11	0,08	0,06	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04	0,04
ICT canola emissions [kg CO ₂ /tkm]	0,07	0,06	0,06	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05	0,05
Total annual tkm transported	2,2E+12	2,5E+12	2,6E+12	2,8E+12	2,8E+12	3,2E+12	3,4E+12	3,7E+12	3,9E+12	4,1E+12	4,6E+12	5,2E+12	5,8E+12
BET annual tkm transported	0,0E+00	1,6E+10	1,1E+11	4,5E+11	8,9E+11	1,4E+12	1,6E+12	2,0E+12	2,3E+12	2,8E+12	3,4E+12	3,9E+12	4,3E+12
ICT annual tkm transported	2,2E+12	2,5E+12	2,5E+12	2,3E+12	1,9E+12	1,8E+12	1,8E+12	1,7E+12	1,6E+12	1,3E+12	1,3E+12	1,3E+12	1,4E+12
Model IAM [kg CO ₂ /year]	218,5	213,9	206,2	206,3	182,2	186,6	179,7	189,9	190,4	189,1	204,3	229,5	251,2
Electric based scenario [Mt CO ₂ /year]	326,1	277,1	213,4	156,0	121,7	128,6	124,0	136,6	143,0	152,8	171,3	194,4	214,3
Biofuel based scenario [Mt CO ₂ /year]	149,9	149,0	145,8	153,4	152,4	171,7	174,8	193,0	202,5	216,1	240,8	272,4	299,9
Diesel based scenario [Mt CO ₂ /year]	218,5	213,4	205,9	216,1	214,3	240,5	243,9	269,3	282,6	301,6	336,1	380,2	418,6

3.2. Human health and metal depletion results for EU and China regions

Table S37. Human health endpoint indicator for China and European Union region scenarios.

HH total [Dalys/tkm]	China region CP			European Union region CP		
	Biofuel	Diesel	Electricity	Biofuel	Diesel	Electricity
ICEV2020	3.6E-07	3.0E-07	7.4E-07	3.6E-07	3.0E-07	6.0E-07
ICEV2025	3.3E-07	2.5E-07	5.7E-07	3.3E-07	2.5E-07	4.6E-07
ICEV2030	3.1E-07	2.4E-07	4.8E-07	3.1E-07	2.4E-07	3.9E-07
ICEV2035	3.0E-07	2.4E-07	4.2E-07	3.0E-07	2.4E-07	3.4E-07
ICEV2040	3.0E-07	2.4E-07	3.8E-07	3.0E-07	2.4E-07	3.1E-07
ICEV2045	3.0E-07	2.3E-07	3.5E-07	2.9E-07	2.3E-07	2.9E-07
ICEV2050	2.9E-07	2.3E-07	3.2E-07	2.9E-07	2.3E-07	2.7E-07

Table S38. Metal depletion endpoint indicator for China and European Union region scenarios.

SOP [kg Cu/tkm]	CN CP			EU CP		
	Biofuel	Diesel	Electricity	Biofuel	Diesel	Electricity
ICEV2020	8.3E-04	7.9E-04	1.0E-02	8.3E-04	7.9E-04	1.0E-02
ICEV2025	9.3E-04	8.0E-04	7.3E-03	9.3E-04	8.0E-04	7.4E-03
ICEV2030	9.3E-04	8.1E-04	5.7E-03	9.3E-04	8.1E-04	5.7E-03
ICEV2035	9.1E-04	8.0E-04	4.9E-03	9.1E-04	8.0E-04	5.0E-03
ICEV2040	9.0E-04	7.9E-04	4.4E-03	9.0E-04	7.9E-04	4.4E-03
ICEV2045	8.8E-04	7.8E-04	4.0E-03	8.8E-04	7.8E-04	4.0E-03
ICEV2050	8.7E-04	7.6E-04	3.4E-03	8.7E-04	7.6E-04	3.4E-03

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