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GHG sustainability compliance of rapeseed-based biofuels produced in a Danish multi-output biorefinery system

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Abstract

Biofuels are likely to play an increasingly important role in the transportation sector in the coming decades. To ensure the sustainability of the biofuel chain, regulatory criteria and reduction targets for greenhouse gases (GHG) emissions have been defined in different legislative frameworks (e.g. the European Renewable Energy Directive, RED). The provided calculation methods, however, leave room for interpretation regarding methodological choices, which could significantly affect the resulting emission factors. In this study, GHG reduction factors for a range of biofuels produced in a Danish biorefinery system were determined using five different emission allocation principles. The results show that emission savings ranged from -34 % to 71 %, indicating the need for a better definition of regulatory calculation principles. The calculated emission factors differed significantly from default values provided in the literature, suggesting that case-specific local conditions should be taken into consideration. A more holistic LCA-based approach proved useful in overcoming some of the issues inherent in the regulatory allocation principles. On this basis, indirect land use change (ILUC) emissions were shown to have the same magnitude as the direct emissions, thus indicating that the overall system should be included when assessing biofuel sustainability criteria.

Keywords: GHG compliance, biofuel, sustainability index, allocation, uncertainty

1. Introduction

In the coming decades, secure energy supply will be challenged by economic factors and technological constraints [1]. To overcome the problem, a mix of different technical and managerial solutions will be needed [2], among which energy-crops-based biofuels should play a relevant role for the transportation sector [3,4]. The development of the biofuel market has been supported and regulated in several countries, in some cases through the establishment of mandatory targets for the use of biofuels as components in road transportation fuels. For example, in the United States, the Energy Independence and Security Act (EISA) of 2007 [5] sets targets for increasing the production of biofuels which will be blended with fossil gasoline and diesel. The EU Renewable Energy Directive (RED) mandates an obligatory 10% minimum target for the share of energy from renewable sources in transportation, which should be implemented by member states by 2020 [6]. This EU biofuel mandate is expected to induce a burgeoning bioenergy trade in the EU [2] as well as the consequent increased production of bioenergy crops both within and outside the EU [7]. The quick growth in energy crop production has raised a number of concerns about the sustainability of biofuels [e.g. 8–10], many of which call for new policies to regulate the industry [2].

To prevent adverse environmental effects, different policy guidelines [e.g. 6, 11] have introduced sustainability criteria for biofuels traded on the market. Within some of these regulatory schemes, transport fuel providers must report/certify the sustainability compliance of fuels [12]. Life cycle thinking (LCT) is typically employed as part of sustainability certification, in order to show that biofuels have lower environmental pressure in comparison to fossil fuel comparators.

Calculation procedures for assessing sustainability are included in several policy directives, and in some cases they are complemented by examples and default values for a number of emission factors. However, and despite such attempts at simplification, the abovementioned sustainability criteria can still arguably be used for assessing the sustainability of biofuels, the reasons for which are multiple. Firstly, directives (and included calculation procedures) leave room for the individual interpretation of some of the key aspects involved in the sustainability compliance calculations. This means that, for example, even within the same calculation method, different choices are possible, providing results conform to the case-to-case need of the certifier. Secondly, not all the directives require the inclusion of indirect land use change (ILUC) effects induced by the increased production of biomass and bioenergy crops [13], despite environmental concerns associated with ILUC effects [e.g. 4,14]. Thirdly, the calculation procedures do not require that they report any uncertainty associated with results calculated within the certification. This deficiency does not put pressure on transparency or stimulate the discussion on what further research should focus on in terms of data collection and modelling.

The objective of this study was to quantify the relevance of some of the issues encountered when calculating the greenhouse gases (GHG) sustainability compliance of rapeseed-based biofuels produced within a Danish biorefinery system. A biorefinery is herein treated as an integrated system, in which multiple processes are employed for biomass refining and several products are delivered, thereby enhancing the sustainability of biomass use [15]. The EU Renewable Energy Directive (RED), which represents the legislative framework for Danish biofuels, is used as an illustrative example. The objective is achieved by: 1) calculating the sustainability index of biofuel products according to the RED; 2) showing how choices and different assumptions made within and accordingly to the RED can provide different results; 3) estimating the uncertainty associated with the results and 4) including impacts related to ILUC effects, to demonstrate their relevance in comparison to the biofuel production phase.

2. Sustainability of biofuels in current legislation

2.1. Sustainability criteria for biofuels

Sustainability criteria provide the basis for certifying biofuel sustainability. Several certification schemes exist internationally, often developed in connection with or as a response to the implementation of renewable energy strategies and targets. These sustainability criteria are based typically on GHG emission factors (EFs), expressing, for instance, the amount (g MJ^{-1}) of CO_2eq emitted per MJ of fuel. A comprehensive overview of the existing criteria is provided by Scarlat and Dallemand [13], who note that these schemes have different natures and objectives: some address the sustainability of renewable energy in general, whereas others are more specific and cover, for instance, only the agricultural stage of bioenergy crops. These certification criteria can be distinguished further between those having legal binding – e.g. the RTFO, the Dutch NTA 8080, the US Renewable Fuel Standard (RFS), the Californian low-carbon fuel standard (LCFS), the EU-RED – and those having a sustainability promotion nature, for example the Global Bioenergy Partnership (GBEP), the International Sustainability and Carbon Certification (ISCC), the Roundtable for Responsible Soy Production (RTRS), the Better Sugarcane Initiative (BSI), the Roundtable on Sustainable Biofuels (RSB), the Roundtable on Sustainable Palm Oil (RSPO) and the Council on Sustainable Biomass Production (CSBP). As a result of differences in calculation methodologies, the abovementioned criteria may provide contrasting results (as shown by e.g. Wardenaar et al. [12]; Börjesson and Tufvesson [16]; Hennecke et al. [17]), thus highlighting how difficult it is to develop generic calculation procedures valid for all renewable energy sources [13]. Differences between criteria can be found in default values for fossil comparators, the allocation method and the inclusion/exclusion of ILUC effects, among others, as illustratively presented in Table 1. All criteria presented in Table 1 allow for the use of default data when specific data are not available, while uncertainty analysis is not required (only RFS recommends uncertainty analysis, albeit solely on the ILUC part).

Table 1 – Examples of differences in methodol	ogical choices in	selected sustainabil	ty criteria for	biofuels. Based on			
Wardenaar et al. [12], Scarlat and Dallemand [2	13], Hennecke e	t al. [17]; Renewable	Fuels Agency	[52]; United States			
Environmental Protection Agency [53]; California Environmental Protection Agency [54].							
			CEC (CAL)				

	RED (EU)	RTFO (UK)	RFS (US)	LCFS (CAL)	RSB (CH)	
Allocation method	Energy	'Restricted'	Substitution	Substitution	Substituion +	
		substitution	substitution		+ energy +	
		+ economy			economy	
Use of default data	Yes	Yes	Yes	Yes	Yes	
Zero-burden for by-products	Yes	Yes	No	No	No	
Inclusion of ILUC effects	No	Yes	Yes	Yes	Yes	
Uncertainty analysis	No	No	For ILUC	No	No	

2.2. The EU Renewable Energy Directive (RED) – a brief introduction

The RED includes sustainability criteria aiming at reducing biofuel production GHG emissions compared to a fossil fuel comparator, as well as protecting biodiversity and land quality [6]. While the latter two issues are defined more from a qualitative point of view, Annex V of the RED provides a methodology (see Supporting Information) for assessing the GHG sustainability index E [6], while EC [18] supplements additional explanations which are useful for calculating in practice. The RED establishes increasing GHG emission targets for biofuels and bioliquids: the minimum saving is currently set at 35%, while it will become 50% and 60% in 2017 and 2018, respectively. In order to calculate E, energy content shall be used for allocating upstream emissions in multi-output processes, whereby a zero-burden approach is assumed for handling by-products and waste, while GHG emissions associated with ILUC

effects will not be included. In addition, the RED provides default values for some of the factors, which are used when actual values are not available. For the cultivation stage of specific biomasses, calculation of emission factors is mandated to individual state member and should reflect geographical conditions. The relevance that such a choice as well as parameter uncertainty may have on the estimation of E, however, is not addressed in this instance. If on the one hand the calculation methodology is simplified to ensure usability, on the other hand some inconsistencies may open up. For example, Buratti et al. [19] reported significant differences between calculated values and default values as a consequence of assumptions regarding rapeseed yield. Some of these issues are tested herein, as described in the following section.

3. Materials and methods

3.1. Test hypothesis

Based on some of the issues and inconsistencies highlighted in the introduction (see Section 1), the following hypotheses being tested within the RED framework were formulated:

- The use of energy as a sole allocation criterion can provide differing results. In fact, in the case of multi-output processes such as biorefinery systems, different options are available for distributing upstream GHG emissions to individual output fuels, even within the same allocation criteria. The allocation can be achieved by: 1) assigning total upstream emissions to the final fuels (biorefinery output) according to their energy content; 2) dividing emissions at each sub-process level according to the energy content of individual sub-process outputs and 3) excluding some of the more dependent products from the calculation, based on, for example, the zero-burden assumption.
- The zero-burden assumption for by-products and waste whose production is not aimed at [18] provides conflicting results. The reason may be twofold: 1) the zero-burden assumption contrasts with the concept of an integrated biorefinery, whose development is supposedly supported by the RED, and 2) some biofuels from residues and co-products falling under the zero-burden assumption might fulfil the sustainability criteria, while their production might not take place because determining products (i.e. biodiesel) do not fulfil the sustainability criteria.
- The use of default values for some of the calculation parameters may provide incorrect results for specific local conditions.
- Uncertainty associated with the input values may be substantial, meaning that clear conclusions on the sustainability of individual biofuels cannot be drawn. The RED does not mandate carrying out uncertainty analysis when calculating E.
- The sustainability of biofuels is questionable once ILUC effects are accounted for. The RED does not include GHG emissions from ILUC effects, despite literature reports that their magnitude may be comparable to emissions from the remaining production chain.

3.2. Biorefinery system description

A biorefinery scheme is used for the conversion of rapeseed into a range of biofuel products (Figure 1), and the integrated concept makes use of both seeds and straw. Rape oil is extracted from seeds by means of mechanical and enzymatic treatment, with rape meal as a by-product. Biodiesel is produced from rape oil through enzymatic transesterification, with methanol as a base-reactant, and obtaining glycerine as by-product. Rape straw is collected and hydrothermally pre-treated to obtain a solid fraction, sent to a simultaneous saccharification and fermentation (SSF) unit (followed by distillation and filtration) for ethanol production, and a liquid fraction sent to a dark fermentation (DF) unit for the production of hydrogen. Effluents from both the SSF and the DF unit are then routed to an anaerobic digestion (AD) unit, for further energy recovery. The digestate produced from AD is used on agricultural land as fertiliser. Energy input (i.e. electricity, heat, fuel) is needed in all processing steps. Mass and energy balances of the biorefinery system are reported in the Supporting Information, together with an

overview of relevant inventory data. A more detailed description of the system, of the inventory data modelling and mass/energy balances is reported in Boldrin et al. [20], while the energy recovered in different products from 1 Mg of rapeseed (i.e. 20 GJ) is summarised in Table 2.

The biorefinery is designed to be flexible, meaning that the chain can be adjusted to prioritise the production of individual co-products according to market demand. These biorefinery co-products can be used for different purposes. In some cases, several options may be possible for individual coproducts [20]. For biofuels, the substituted fossil fuels to compare with were chosen from among the options provided [6]: biodiesel with diesel, ethanol with gasoline, hydrogen with industrial hydrogen, methane with natural gas and lignin with lignite. Glycerin was assumed to be used in industrial boilers for the production of heat and electricity, thereby substituting for natural gas [20]. Rape meal was assumed to be used as animal fodder, thus displacing the production and use of soybean and barley meals; the replacement is modelled by balancing the amount of proteins and carbohydrates supplied with the meal [20]. The digestate used on agricultural land was assumed to replace mineral fertilisers, in an amount proportional to the N, P, K content in the digestate, as reported in Boldrin et al. [20].



Figure 1 - The rapeseed-based biorefinery system. Dotted products represent the "avoided products" offset by the coproducts from the biorefinery; ILUC impact occurs as a consequence of increased use of land in Denmark for rapeseed cultivation.

Inventory data for the production of rapeseed in Denmark, soy in Brazil and barley in Canada were taken from Schmidt [21]. The inventories were developed by taking into consideration local conditions (e.g. yield, soil and climate) and assuming intensified cultivation driven by increased demand for crops on the market. The inventories include the provision and use of energy and materials, emissions occurring during the agricultural stage and the transportation of material and products to and

from agricultural land. In the inventories, field emissions of N_2O from the use of N fertilizers were estimated based on the IPCC emission model [22].

3.3. Allocation criteria

Accounting for GHG emissions is done in order to document that biofuels have emission factors lower than the fossil fuels they substitute (known as 'fossil fuel comparators'). Within the RED, overall GHG emissions from the biorefinery system were allocated to the products on an energy basis. Five calculation criteria, all acceptable within the RED framework, were identified for calculating $E_{o,f}$ (emission factor *E* for output *o*,*f*).

(a) Total biorefinery GHG emissions were shared among all biofuel co-products (i.e. rape meal and digestate are excluded), using the following factor α_i :

$$\alpha_i = \frac{En_{o,f}}{En_{tot}}$$
 (Equation 3)

where *En*_{o,f} is the energy contained in product *o*,*f*, and *En*_{tot} is the total energy output of the biorefinery.

(b) $E_{o,f}$ was calculated as in criterion (a), including all co-products (i.e. biofuels, rape meal and digestate) in the system.

(c) The allocation was performed at the process level, meaning that for each sub-process, emissions related to inputs into that process were allocated on an energy basis to the process's outputs.

(d) Lignin and glycerine are used internally in the biorefinery for energy production, and they are thus no longer listed among the biofuels. When calculating $E_{o,f}$, emissions avoided from electricity delivered to the grid were subtracted according to Equation 1, using an energy allocation as in criterion *a*. Emission factors e_{ee} for the avoided electricity were assumed to be those when "electricity was generated in a power plant using the same fuel as the cogeneration unit" [6]. An e_{ee} of 23 g kWh⁻¹ of CO₂eq was assumed for lignin, corresponding to an emission for biomass-based electricity production in Nordic countries [23]. Glycerine can be combusted in an industrial boiler for the production of thermal energy (e.g. steam, hot water, etc.). Despite the use of glycerine could displace the use of natural gas [20], no emissions should be allocated to heat, according to EC [18].

(e) Production of rapeseed residues (i.e. straw) was assumed burden-free (i.e. zero-burden). Thus, GHG emissions from rapeseed production were all allocated, according to criterion b, to biodiesel, glycerine and rape meal, while emission factors for ethanol, biohydrogen, methane and lignin only included burdens associated with inputs of materials and energy into the sub-processes.

Allocation factors calculated according to criteria a to e are presented in Table 2, where standard deviations were assumed as calculated with STAN [24] during mass and energy balances reconciliation (details in Boldrin et al. [20]). Additional details about allocation factors calculation are provided in Supplementary Information. Relevant differences among criteria can be seen, with biodiesel having a share of the overall emissions, ranging between 41 % and 79 %. In particular, a large difference is due to including (i.e. criteria b, c, e) or excluding (i.e. criteria a, d) the by-products, which may thus represent a critical assumption in the calculation methodology. Criterion e, calculated assuming zeroburden, does not deviate significantly from criteria c and d, possibly because emissions from energy-intensive processes in the production of ethanol counterbalance the exclusion of rapeseed provision.

In addition to the above presented criteria, the overall GHG emission for the system is calculated, in order to estimate the relative GHG reduction compared to the conventional (fossil) system. Modelling is done based on the LCA approach and therefore includes both fossil comparators as avoided products and ILUC emissions related to the provision of rapeseed. As explained in Section 3.2, the ILUC effects result from both increased rapeseed production in Denmark (affecting agricultural

production) and the increased production of rape meal, with the consequently lower importation of other meal (e.g. soybean and barley).

	Output	Energy	Allocation factors (%)						
		(MJ)	а	b	С	d	е		
Biofuels	Biodiesel	7750±120	68.2±1.4	45.1±0.9	41.3±0.8	78.7±1.6	49.9±5.9		
	Bioethanol	785±20	6.9±0.2	4.6±0.1	5.3±0.1	8.0±0.2	2.7±0.4		
	Biohydrogen	110±2.8	1.0±0.03	0.6±0.02	1.1±0.03	1.1±0.03	0.4±0.06		
	Biomethane	1200±30	10.5±0.3	7.0±0.2	10.3±0.3	12.2±0.3	4.1±0.7		
	Lignin	1120±28	9.9±0.3	6.5±0.2	7.6±0.2		3.9±0.6		
	Glycerin	400±10	3.5±0.1	2.3±0.06	2.1±0.06		2.6±0.3		
By-	Rape meal	5480±130		31.8±	29.3±		35.2±4.2		
products	Digestate	356±8.9		2.1±	3.0±		1.2±0.2		
Total		17200	100	100	100	100	100		

Table 2 – Energy content (referring to 1 Mgww rapeseed, i.e. 20006 MJ) of outputs from the biorefinery (from Boldrin et al. [20]) and energy-based allocation factors used in the calculation (totals may not add up because of rounding off).

3.4. Calculation setup and uncertainty analysis

LCA modelling was facilitated in SimaPro 7.3.3 LCA software [25]. As explained in detail in the next section, inventory data for the biorefinery system were partly provided by project partners, and then complemented with inventory data from ECOINVENT 2.2 [26]. Marginal electricity production was assumed from coal [20], while the commissioning and decommissioning of the infrastructure were excluded, as stated in EC [6]. Emission factors for the fossil fuel comparators were chosen according to EC [6]. The global warming potential of biogenic CO₂ emitted from biofuel combustion was assumed neutral (i.e. GWP=0) [27]. Uncertainty related to biofuel emission factors was estimated by means of Monte Carlo analysis (1000 simulations), which was applied to both the allocation factors and the process parameters. When specific data were not available, ECOINVENT default uncertainty values (reported in Supporting Information) were used for process parameters [26]. For some of the most relevant parameters identified during the first iteration, uncertainty was defined according to the following considerations:

- Rapeseed yield varies from year to year and is steadily increasing in Denmark because of improved land management and operations. The additional demand for rapeseed is assumed to be satisfied by both agricultural intensification (40% of the amount) and expansion (60%), as explained in Schmidt [28]. The estimation was based on FAPRI modelling, with an annual estimated increase of 1.01% in rapeseed yield and 1.52% in cultivated area (in line with Scarlat et al. [29]). The rapeseed yield was defined as being distributed uniformly between 3.26 Mg ha⁻¹ (as suggested in Schmidt [21]) and 3.57 Mg ha⁻¹, as reported in Statistics Denmark for 2010 (see Supporting Information).
- Relevant N₂O emissions occur during nitric acid production required for inorganic N-fertiliser production. These emissions can have a significant impact on the GHG balance of biofuels [30], while their magnitude is particularly variable and depends on the production technology. The emission factor for N₂O was defined as being distributed uniformly between 8.39 kg kg⁻¹ of acid (ECOINVENT process "Calcium ammonium nitrate, as N, at regional storehouse/RER U") and 1.26 kg kg⁻¹ of acid [31], as suggested by Schmidt [21].

- Energy consumption during field operation was defined as being distributed uniformly between 3,612 MJ ha⁻¹ [21] and 3,090 MJ ha⁻¹ [32], both datasets being estimated for the cultivation of rapeseed in Danish conditions.
- Transportation distance for rapeseed was distributed uniformly between data reported in Schmidt
 [21] (i.e. 366 t·km in a medium-sized truck and 1,942 t·km in a large truck) and Sambra et al. [32]
 (i.e. 250 t·km and 1,350 t·km in medium and large trucks), in order to take into consideration the optimised logistics of rapeseed.

3.5. Indirect land use changes (ILUC)

In response to the growing demand for biofuels, increased production of terrestrial energy crops can be achieved by intensifying existing production or by expanding cropland into uncultivated areas [33]. As a result of global market-driven mechanisms, the expansion of cropland can occur in regions geographically very distant from the place where energy crops and biofuels are produced. When cropland is expanded into uncultivated land, the so-called ILUC effects may occur as a consequence of activities (e.g. vegetation clearing, soil tilling, etc.) involved in land conversion. The magnitude of detrimental ILUC effects, in some cases, can be comparable to the benefits related to the avoided use of fossil fuels [8,9], thus making the sustainability of biofuels highly questionable. The identification of ILUC effects requires complex agro-economic modelling, which can provide very different results depending on certain specific assumptions [34]. An overview of ILUC-GHG emission factors for European biodiesel production from rapeseed is provided in Table 3, which includes only studies providing an estimation of ILUC emissions specifically for rapeseed biodiesel. From Table 3, an emission factor for CO_2eq in the range 33-67 g MJ^{-1} was identified and used for comparing direct and indirect GHG emissions occurring during biodiesel production.

Fuel	Crop	Unit	Value	Region	Notes	Reference
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	52	Europe	Mirage model, TH= 20 yr	[7]
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	33	Europe	Arable Land, 25% iLUC	[3]
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	67	Europe	Arable Land, 50% iLUC	[3]
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	54	Europe	Mirage model - TH= 20 yr	[55]
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	45	Europe	TH= 20 yr	[56]
Diesel	Rapeseed	g MJ ⁻¹ of CO ₂ eq	52-57	Europe	From IFPRI, TH=20 yr	[57]
Diesel	Mix	g MJ ⁻¹ of CO ₂ eq	45	Europe	TH= 20 yr	[56]
Diesel	Mix	g MJ ⁻¹ of CO ₂ eq	57	Europe	GTAP model, TH= 30 yr	[58]

Table 3 – Overview	of ILUC-GHG	factors related to	rapeseed-based	biodiesel	production i	n Europe.

TH: time horizon

As presented by Schmidt [21], an alternative estimation of ILUC emissions can be based on consequential LCA modelling for identifying the marginal agricultural production of rapeseed in Denmark. Conversely from the agro-economic models listed in Table 3, this method assumes full elasticity of supply, in order to recognise that the increased production of rapeseed in Denmark would displace Danish barley production, which would be replaced subsequently with barley imported from Canada, where barley growing would be increased [28]. It is in fact predicted that Canada will cover the largest increase in barley production in the short to medium term [35] – for an additional 1 Mg of rapeseed produced in Denmark, Canada will grow an additional 780-880 kg. Using the LCI presented in Schmidt [21] for Canadian barley production, ILUC emissions (in CO_2eq) may be in the order of 503±37 kg Mg⁻¹ of rapeseed, or 29±2 kg MJ⁻¹ to 79±6 kg MJ⁻¹ of fuel depending on whether the ILUC is allocated to all products (criteria *b*) or only to the biofuels (criteria *a*), respectively.

4. Results

Overall GHG emissions from the production of biofuels from 1 Mg of rapeseed were estimated at 981 ± 74 kg of CO₂eq for all *a*, *b*, *c* and *e* criteria, while it was 887 ± 73 kg of CO₂eq for criterion *d* where savings from electricity production are internalised. Emission factors calculated according to different criteria are presented in Table 4, together with emission savings estimated in comparison to the respective fossil fuel comparators. The results are then visualised in Figure 2, where they are compared to current (i.e. year 2010) and future (i.e. years 2017 and 2018) emission saving targets (dotted lines). It is evident that the assessed criteria provide rather different values for E, ranging from positive (83%) to negative (-34%). In general, when taking into consideration uncertainties, it is understood that most of the biofuels fulfil or are close to the sustainability target when using criteria *b* and *e* for the calculation of E. Conversely, criteria *a* and *d* estimate emission savings which do not comply with RED targets. Within criterion *c* (and partly also criteria *b* and *e*), some of the products from the biorefinery would comply with RED targets, while others would not do so.

From an energy point of view, biodiesel is the main output of the biorefinery system. Results in Table 4 and Figure 2 show that, for some of the allocation criteria, biodiesel complies with RED emission saving targets. These figures, based on the current technological level for biofuel production in Denmark, however, are quite far from the targets set for years 2017 and 2018. Despite fast technological development, which allows for predicting that relevant improvements will be obtained in the near future [36,37], major efforts are needed to improve the performance of the production chain and to fulfil future stricter emission targets.

The largest share of GHG emissions is the result of rapeseed production and transportation (about 773±69 kg of CO₂eq), while the remainder (~10-20%) originates from processing. Rapeseed production is thus the stage at which improvements could and should be achieved. As shown in the contribution analysis (see Supporting Information), the major contributors to GHG emissions during rapeseed production are connected to the use of inorganic fertilisers (especially N, which results in large N₂O emissions), field operations and rapeseed/straw transportation. The critical contribution of N fertiliser confirms previous findings [e.g. [17,19,38–40]. In this respect, the GHG footprint of inorganic fertilisers could be improved through the use of BAT production systems [21], wherein fugitive N₂O emissions are reduced by employing abatement measures. Field operations could also be improved through optimised management and more efficient machinery, while logistics could be improved significantly if transportation distances were shortened. The latter could be achieved with more evenly distributed instalments of smaller biorefinery sites on the territory; previous studies indicate that the scale of the biorefinery plant may not have a significant influence on emission factors [41]. The use of pesticides during rapeseed production does not make a major contribution to GHG emissions. Nonetheless, pesticides could damage the environment and human health, and have adverse effects on biodiversity when reaching non-targeted life forms [42]. Their use should therefore be evaluated from a broader sustainability perspective (as defined in the RED), where effects on biodiversity and land-use are also addressed alongside GHG emissions.

Biofuel processing accounts for 10-20% of GHG emissions throughout the production chain. Important improvements to the overall sustainability of this chain could thus also be achieved through the further optimisation of the processing phase. In particular, as highlighted by Boldrin et al. [20], minimising process losses and enhancing conversion efficiencies could be key aspects on which to focus in future research. Conversion processes with significant material and energy losses, for example, are hydrothermal pre-treatment, dark fermentation, AD and SSF. Energy-intensive processes, where a more efficient conversion may be sought, include enzymatic transesterification, hydrothermal pre-treatment and dark fermentation.

		Biodiesel	Bioethanol	Biohydrogen	Biomethane	Lignin	Glycerin	Rape meal	Digestate
Emission factor E	Criteria	86.3±6.9	86.6±7.4	86.4±7.6	86.0±7.3	86.4±7.4	86.5±7.4		
(g MJ ⁻¹ of CO ₂ eq)	а								
	Criteria	57.2±4.4	57.1±4.9	57.1±4.8	57.1±4.8	57.1±4.8	57.0±4.8	56.9±4.6	57.0±4.8
	b								
	Criteria	52.5±4.3	65.8±5.7	100.6±8.2	83.6±7.2	66.7±5.7	52.3±4.3	52.4±4.4	83.8±7.1
	С								
	Criteria	89.9±7.6	89.9±8.1	90.2±8.2	90.4±7.9				
	d								
	Criteria	62.7±5.8	34.0±4.6	33.7±4.5	33.6±4.5	33.8±4.7	63.1±5.7	63.3±5.8	33.6±4.6
	е								
	Criteria	1.5±7.9	-3.3±8.8	1.1±8.7	-27.2±10.8	56.7±3.7	0.8±8.3		
GHG emission	a	04 7 . F 0	24.0.5.0	246.55	45 6 3 4	74.0.0.4	245.56		
saving S (%)	Criteria	34.7±5.0	31.9±5.8	34.6±5.5	15.6±7.1	/1.2±2.4	34.5±5.6		
	D	4.0	21 416 9	15 210 4	22 7 10 6	66 212 0	20.015.0		
	Criteria	4.9	21.4±0.8	-15.3±9.4	-23.7±10.0	00.3±2.9	39.9±5.0		
	L Critoria	2 6±0 6	7 2+0 7	2 2 1 0 1	22 7+11 7				
	d	-2.010.0	-7.319.7	-3.319.4	-55.7±11.7				
	Criteria	28 5+6 6	59 4+5 5	61 5+5 2	50 2+6 7	83 2+2 3	27 6+6 9		
	e	20.520.0	55.125.5	01.3_3.2	50.220.7	00.222.0	27.020.5		
Fossil fuel	-	Diesel	Gasoline	Hydrogen	Natural gas	Generic	Generic		
comparator (g MJ ⁻¹ of CO ₂ eq)		87.6*	83.8**	87.3*	67.6*	87***	87***		

Table 4 – Emission factors (g MJ-1 of CO2eq) and GHG emission savings (%) for biofuels produced within the rapeseed-based Danish biorefinery system, and fossil fuel comparators. Errors are standard deviations calculated after Monte Carlo analysis.

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* Source: [58] ** Source: [59]

*** Source: [60]



Figure 2 - GHG emission savings (%) for rapeseed-based biofuels produced within the Danish biorefinery system calculated according to different allocation criteria. The horizontal dotted lines represent emission saving targets set by RED [6]. The error bars are standard deviations calculated after Monte Carlo analysis (1000 simulations).

5. Discussion

5.1. Allocation criteria, system boundary and zero-burden

The results show considerable variability in values of E, as a consequence of how the energy-based allocation procedure is interpreted. In our opinion, this issue should be better addressed in the RED (and ultimately in other pieces of legislation), as it could create confusion from an industrial standpoint during the reporting process. As expected, criteria where emissions are allocated over fewer products provide, in general, higher emission factors and lower emission savings. This is the case, for example, for criteria *a* and *d*, which mostly result in negative emission savings (i.e. emissions from biofuels are higher than fossil comparators).

The way a system boundary is defined may also significantly affect the results. For example, in criterion *d* it was assumed that lignin and glycerine were used internally in the biorefinery for energy production. However, circumvented electricity production – due thanks to delivered electricity – has a small emission factor (i.e. 23 g kWh⁻¹ of CO₂eq for electricity from lignin, corresponding to an emission for biomass-based electricity production in Nordic countries), meaning that much smaller benefits are credited to the system compared to other criteria, where avoided electricity is credited with an emission factor corresponding to the regional mix.

Criterion *b*, criterion *c* and criterion *e* provide quite controversial results, as some biorefinery products would comply with RED targets, while others would not do so. This could indeed result in problematic management situations, where some of the biofuels from the same system could be marketed, while others could not and may need to be discarded as waste. A vicious circle would then follow, as waste product would be excluded from RED calculations, thereby worsening emission factors for other biofuels when the calculation is iterated. This is particularly true for criterion *e*, where the zero-burden was assumed for residues: biofuels produced from straw show large emission savings, while biodiesel does not fulfil GHG sustainability criteria. This could be rather problematic from an industrial perspective during reporting procedures [43].

The abovementioned issues, i.e. allocation criteria, system boundaries and the zero-burden approach, suggest that the calculation methodology should probably be revised so that emission factors could be estimated unequivocally for different biofuels.

5.2. Use of default values

In Table 4, the emission factor of CO₂eq for biodiesel – the main energetic output – is 52-90 g MJ⁻¹. This value is significantly higher than both default values provided in the RED (i.e. 46 and 52 g MJ⁻¹ of CO₂eg for typical and default values respectively) and values previously reported in the literature (e.g. 40 and 55-56 g MJ^{-1} of CO₂eq in Milazzo et al. [44] and Buratti et al. [19], respectively), thereby explaining the lower emission savings for Danish rapeseed biodiesel compared to default values in the RED (i.e. 38-45%) and DG Energy [36] (i.e. 40-60%). In particular, for the rapeseed produced in Denmark, the GHG emissions (i.e. CO2eq) were estimated to be 68±6 or 45±4 g MJ⁻¹ of RME, depending on whether rape meal and digestate were excluded or included in the energy allocation. Both values are significantly higher than the default values (i.e. 23-27 g MJ^{-1} of CO_2eq) calculated for Danish conditions at the NUTS 2 level by Elsgaard et al. [45]. The difference is possibly due to the fact that the present study, compared to Elsgaard et al. [45], makes use of a higher energy allocation factor to biodiesel, it includes transportation of rapeseed, and it assumes intensified cultivations with subsequent larger consumption of inorganic fertilizers. This highlights the necessity for scoping the assessment to local conditions (e.g. geography, technology, management, etc.), as highlighted clearly by Soimakallio and Koponen [46] and Buratti et al. [19]. For example, a relevant factor for the GHG balance of rapeseed production is yield, which is determined largely by geographical (e.g. soil type) and climatic conditions [47], as well as the technological level of the region in question. Yield can vary significantly from country-to-country and from year-to-year, thereby suggesting that average emission factors may not be

representative of local conditions. Other factors associated with the local climate, for example, are the use of fertilisers and related GHG emissions (in particular N₂O) during cultivation.

While acknowledging that default values are useful in many cases when time-consuming and costly data collection is not feasible, we also suggest that default emission factors could be provided in the future for specific geographical regions, thereby allowing a more accurate estimation of emission savings.

5.3. Results uncertainty

The results presented in Table 4 and Figure 2 show that some of the values of E are associated with significant uncertainty, owing to several factors, including uncertainties regarding mass and energy balances, rapeseed yield, transportation distances and emission factors for some of the material and energy inputs into the system. In some cases this uncertainty makes it difficult to determine univocally whether individual biofuels comply with GHG sustainability criteria (see, for example, Figure 2). Similarly to what was concluded by Mullins et al. [48] and Rajagopal & Plevin [49], it seems clear that uncertainty cannot and should not be neglected, while providing 'precise' figures for E may indeed be misleading.

Besides using accurate and case-specific inventory data, it is thus recommended that future studies should include estimations of the uncertainty associated with the results, thereby providing more transparent conclusions. In this respect, it is additionally suggested that procedures for uncertainty analysis should be provided in related legislation.

5.4. ILUC emissions and life cycle assessment (LCA)

As shown in Section 3, direct emissions and ILUC emissions are in the same order of magnitude. For the biofuel chain under assessment, direct emissions from the refining process were estimated to be in the order of 34 to 101 g MJ^{-1} of CO_2eq (Table 4). This is in line – albeit at the higher end – with DG Energy [36], where emission factors in the order of 39-93 g MJ^{-1} of CO_2eq are reported. On the other hand, as previously mentioned, ILUC emissions are in the range 29-79 kg MJ^{-1} of CO_2eq . This means that if ILUC emissions are added to direct emission, emission savings would be close to zero or negative (i.e. biofuels result in more emissions than fossil comparators), regardless of the allocation criteria chosen. This indicates that although the magnitude of ILUC emissions should be clarified better, further biorefinery optimisation is needed to ensure that biofuels are sustainable, even with all the uncertainties highlighted so far.

A more precise GHG balance of biofuels was established by running a fully consequential LCA where ILUCs were accounted for as increased production of barley yields in Canada (see section 3.5 for details). The LCA includes avoiding (substituting) the use of fossil fuels, a decrease in the importation of soybean and barley meals as a result of the increased production of rape meal, and the benefits from heat production from glycerine and lignin. The overall GHG (i.e. CO_2eq) balance of the system is estimated to be 156±123 kg Mg⁻¹ of rapeseed, corresponding to an emission factor in the range 9±7 to 14±11 g MJ⁻¹ of CO₂eq. These figures are very close to zero (i.e. no difference between the biofuel and fossil systems) and associated with considerable uncertainty, meaning that a clear conclusion on whether biofuels are more sustainable than fossil fuels (comparators) cannot be drawn. These results conversely highlight that a more holistic approach (e.g. by using an LCA) may be more appropriate when assessing the sustainability of biofuel production.

Using an LCA for assessing the sustainability of biofuels has both advantages and disadvantages. Some of the advantages include 1) circumventing critical assumptions regarding the allocation of emissions to different biofuels; 2) a better understanding and comparison of direct and ILUC emissions; 3) excess heat for heating purposes can be modelled, which represents a key aspect for improving the economical sustainability of integrated biorefineries [50] and 4) a more comprehensive single value is provided for all co-products, thereby avoiding some of the controversial issues highlighted previously (i.e. within the same biorefinery, some biofuels are in compliance, while some are not). Some of the drawbacks of the LCA approach are 1) it significantly increases the complexity of the study, thereby still leaving room for subjective decisions by the LCA practitioner (e.g. avoided processes) and related uncertainty [51] and 2) it does not allow for setting specific targets, as an LCA is better suited to providing relative and comparative figures rather than absolute values.

6. Conclusions

The GHG sustainability index of a range of rapeseed-based biofuels produced in an integrated biorefinery system under development in Denmark was calculated. Emission savings were determined using five different allocation criteria identified within the calculation methodology described in the Renewable Energy Directive (RED), which were then compared to the minimum sustainability targets established for the marketability of biofuels. Significantly different emission savings were obtained depending on the allocation criteria and the system boundary adopted – ranging from -34% to 83%. In some cases, results led to contradictory situations – some of the biofuels fulfilled the targets, while others did not.

The largest contribution to GHG emissions was identified as being related to rapeseed production, which accounts for about 80-90% of the direct emissions. In particular, the use of fertilisers, field machinery and transportation were identified as important hotspots where improvements should be sought, together with a general enhancement of conversion efficiencies in the biorefinery. Some of the calculated values significantly differed from default values provided in some of the methodologies for GHG calculation (e.g. RED), indicating that GHG sustainability compliance should always be verified in relation to local conditions and suggests that regionalised default values may prove to be more useful.

From a policymaking perspective, the present study demonstrated that an unequivocal clarification of the procedure for calculating GHG emission reduction targets should be provided as a matter of urgency, while encouraging the use of case-specific data instead of default values, which may provide largely incorrect results, should also be actioned. Additionally, it is recommended that calculation procedures demand for a systematic uncertainty analysis of the results. Finally, including ILUC emissions proved to be crucial for concluding on the overall environmental sustainability of biofuels.

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