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Cottonseed Oil: a Possible Feedstock Alternative for Renewable Diesel Production

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I wish you all the best.

As Nature works around all obstacles and turns them
in accordance with the laws of Destiny, so can the rational being
craft any impediment into matter for his or her own action.

Marcus Aurelius

Meditations VIII, 35

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Sommario

L’oggetto di questa ricerca è lo studio delle emissioni di gas serra nel ciclo di vita (LCA) del diesel rinnovabile (RD) da olio di semi di cotone. L’interesse verso questo come altri olî alternativi è dettato dalle recenti direttive della UE, particolarmente stringenti per alcuni produttori di RD in quanto implicanti l’eliminazione dell’olio di palma entro il 2030, tra le altre misure contenute; tra i produttori più interessati spicca anche l’Italia, ove l’olio di palma costituisce circa l’80% della materia prima processata dalle sue due bioraffinerie di Marghera e Gela [1]. A differenza di altre materie prime, l’olio di cotone non è stato studiato in modo approfondito in questi termini. Un solo caso studio peer-reviewed è attestato in letteratura, per un impianto nel Brasile rurale; tale impianto non produce RD, ma biodiesel [2]; un solo altro impianto a biodiesel, non peer-reviewed, è stato studiato in uno studio legato a un contesto rurale in India [3]. Questa ricerca va quindi a investigare una materia prima effettivamente poco studiata.

Gli obiettivi di ricerca sono i seguenti: (I) definire l’olio di semi di cotone secondo le direttive UE in materia di biocarburanti, e quantificare l’intensità di riscaldamento globale (GWI) del RD da olio di semi di cotone per valutarne la conformità a tali direttive; (II) valutare il GWI di tale biocarburante a confronto con altri olî d’uso comune, attraverso un LCA che aderisca alle direttive ISO, le più diffuse in questo genere di studî. Tali obiettivi sono stati perseguiti sviluppando un modello con il supporto del software LCA GaBi. Lo sviluppo del modello è stato basato su un caso studio per Israele: da qui, l’olio viene poi trasportato in Italia per la raffinazione; la ricerca è stata sviluppata in collaborazione con l’università Technion di Haifa. Per ciascuno dei due obiettivi, è stato necessario elaborare un modello distinto: il modello EUC copre il primo obiettivo, il modello IS il secondo. Il sistema, in entrambi i casi, comprende: coltivazione, sgranatura, estrazione d’olio in Israele; raffinazione e distribuzione - quest’ultima solo nel modello EUC - in Italia; a collegare questi processi sono stati altresì modellati dei processi di trasporto, via terra o via mare. Il modello IS differisce dallo EUC principalmente per l’inclusione, entro i confini del sistema, dell’industria casearia israeliana, dove ha sede il consumo di seme e farina di cotone; questo implica l’inclusione di una filiera per l’approvvigionamento di integratori alimentari - in questo caso, cereali - che vadano a supplire alla mancanza di seme grezzo causata dall’esportazione d’olio a scopo energetico. È bene sottolineare che la natura del caso studio non consente estensione indiscriminata dei risultati all’infuori dei risultati; se da un lato il modello IS mette in luce le caratteristiche del RD da cotone rispetto ad altri biocarburanti, e i trade-off che il suo ciclo produttivo implica, si ritiene che sia sempre necessaria un’analisi case-specific qualora si voglia pervenire a valori di

GWI per un altro contesto.

Il modello EUC suggerisce che il RD in esame è largamente conforme alle direttive UE. Il suo GWI risulta 16 g_{CO2eq}/MJ di combustibile, e il suo limitato intervallo di variazione suggerisce ampie possibilità di sfruttare l'olio di cotone in conformità con le suddette direttive (il cui limite più stringente ammonta a 33 g_{CO2eq}/MJ). Se confrontato con le più comuni materie prime, il GWI del RD in esame è secondo solo agli olî vegetali esausti in quanto a riduzione dell'impronta di carbonio: gli olî di palma, colza e soia risultano tutti in GWI dalle due alle tre volte più alti rispetto all'olio di cotone. Il modello IS suggerisce che il RD prodotto è meno impattante del diesel fossile, per emissioni dirette. In questo caso, i risultati sono meno univoci, e la scelta dell'integratore alimentare usato nell'industria casearia è la determinante più importante per il GWI finale. Cambiare integratore, infatti, può portare il GWI da 83 a 63 g_{CO2eq}/MJ; anche il trasporto di tali cereali risulta avere un effetto importante sul GWI, con un contributo ad esso fino a 10 g_{CO2eq}/MJ: un'attenta scelta degli integratori può quindi portare a GWI potenzialmente migliori di olî di prima generazione come colza e palma, con GWI tra i 50 ed i 60 g_{CO2eq}/MJ. In aggiunta a ciò, le emissioni da indirect land use change (ILUC) risultano essere più basse di circa 25-30 g_{CO2eq}/MJ rispetto a colza e palma; scegliendo un integratore alimentare che sia carbon-efficient, il GWI inclusivo di ILUC del caso studio risulta 80 g_{CO2eq}/MJ, sotto i 94 del diesel fossile e ben sotto i GWI complessivi stimati per palma e colza.

La liceità dell'uso di olio di cotone per produrre RD potrebbe portare, nel medio termine, ad un suo uso per sostituire l'olio di palma nelle bioraffinerie europee. In alcuni Stati africani in particolare, il seme di cotone è attualmente poco sfruttato: quello dell'Africa potrebbe quindi rappresentare un contesto particolarmente favorevole cui fare affidamento. In questo senso, l'impronta carbonica e l'effettiva creazione di valore sul territorio di un progetto, senza impattare su altri mercati esistenti, dipendono in larga misura da un'accurata analisi del contesto specifico. Risvolti prolifici potrebbero in futuro risultare da ricerca in contesti specifici, ad esempio africani, con un più alto potenziale di olio di Israele, con o senza allevamenti bovini annessi. Altri sviluppi interessanti potrebbero aversi confrontando diversi regimi di approvvigionamento di integratori alimentari, studiando l'impronta carbonica al variare di aspetti quali l'origine geografica e le pratiche agricole. Studi che abbraccino altre categorie d'impatto oltre al cambiamento climatico possono fungere da ulteriore approfondimento in quanto a sostenibilità del biocarburante.

Parole chiave: LCA; biocarburanti; cotone; idroconversione; ILUC.

Abstract

This research studies the life cycle of cottonseed-oil renewable diesel (RD) in its greenhouse-gas emissions impact. Unlike other biofuel feedstocks, cottonseed oil has not been subject to much attention from the scientific community. Interest on alternative feedstocks stems from the latest EU regulations, which are likely to cause a shock in the feedstock supply of RD producers in the region. The single most impacting policy is the complete phase-out of palm oil by 2030, which will have consequences in particular for some EU countries, among which also Italy. Thus, the main goals of this study are as follows: (I) defining cottonseed oil in the framework of EU biofuel regulations and quantitatively assessing the Global Warming Intensity (GWI) of cottonseed-oil RD, in order to check its compliance with said regulations; (II) assessing the GWI of this biofuel in comparison with other feedstocks, in the framework of the most widespread guidelines in matter of Life Cycle Assessment (LCA). These objectives have been pursued by developing an LCA model using GaBi, an LCA software, as platform. Model development was based on a case study for Israel: from here, the oil is shipped to Italy to be refined; the research was developed in collaboration with the Technion, the leading technical university in the country. Two different models were developed, each pursuing one of the two mentioned main goals. The first model suggests that cottonseed-oil RD is largely compliant with EU biofuel regulations. Its baseline GWI of around 16 g_{CO2eq}/MJ and its limited range of variation suggest compliance under a wide range of contexts, since it is well below the EU compliance threshold, which in its strictest form amounts to about 33 g_{CO2eq}/MJ. The second model suggests that this fuel has lower direct emissions than fossil diesel. Here, the range of variation is pronounced, which makes the comparison with other biofuels less univocal. The choice of a particular feed supplement seems to be the most important determinant of the GWI. By merely changing supplement, such value can shift from 83 to 63 g_{CO2eq}/MJ; feed supply chain can also influence the model results by up to about 10 g_{CO2eq}/MJ. Thus, a thorough choice of feed supply seems to lead to GWIs in the range of 50 to 60 g_{CO2eq}/MJ, potentially lower than other first-generation feedstocks such as palm and rapeseed oil. In addition, cottonseed-oil RD appears to prompt massively less indirect land use change (ILUC) than other common first-generation oil feedstocks. The difference is assessed at 25 to 30 g_{CO2eq}/MJ in favour of cottonseed: if a carbon-efficient feed production is chosen, the resulting GWI keeps below fossil diesel, at around 80 g_{CO2eq}/MJ.
Keywords: LCA; biofuels; cottonseed; hydroconversion; ILUC.

Extended abstract

Decarbonising the transport sector is a key challenge that modern climate policies have to address. Policy-makers are well aware that this sector alone represents about a quarter of global Greenhouse Gas (GHG) emissions and that it is lagging behind the other energy sectors: in the last decade, the share of biofuels in global transport energy consumption rose of just one percentage point, from 2% to 3% [4] [5]. The role of policies in this sector is fundamental: the importance of regulations, in fact, is such that biofuel production volumes, in Europe, are almost exclusively explained by the targets set in biofuel mandates, with tax incentives and oil barrel price exerting only secondary effects [6]. In the EU, regulations take the form of directives and implementing acts that set common guidelines and binding targets. The EU Directive 2018/2001, commonly referred to as "RED II", sets a Union target of 14% by 2030 in the share of renewable energy used in the transport sector, while the previous RED set a target of 10% for 2020 [7]. Although electrification and use of alternative fuels will play an increasingly important role in the decarbonisation of transport, the new target is expected to be met primarily via an increase in the share of liquid biofuels [8] [9]. In this context, renewable diesel (RD) is an appealing option: on the one hand, its fuel characteristics are superior to its direct competitor biodiesel (FAME), making it a drop-in biofuel which would be suitable also for the decarbonisation of complex sectors such as aviation [10] [11]; on the other hand, it is based on a rather new but consolidated technology, with a production cost consistently inferior to all other drop-in alternatives [12].

The GHG intensity of RD is primarily influenced by the nature of the feedstock. Kalnes et al. [11] [13] have carried out life-cycle assessment (LCA) studies on soybean oil, rapeseed oil, tallow oil and palm oil; Arvidsson et al. [14] and Uusitalo et al. [15] have studied palm, rapeseed and jatropha oil; Huo et al. [16] carried out an LCA of soybean oil RD by also modelling the hydroconversion reactor section. From these studies, which all adhere to the internationally-recognised ISO LCA guidelines, the Global Warming Intensity (GWI) of RD results in the range of biodiesel GWIs, with direct GHG emission savings always in excess of 40% when compared to fossil diesel. Such studies recognise the importance of co-products in reducing the fuel carbon footprint, and the issue of how to account for these co-products. In order for a biofuel to be compliant with EU directives, a GHG emission reduction threshold must be met: RED II provides guidelines to be followed in the assessment of a biofuel GWI, as well as default GWI values for all the most common biofuel pathways, based on research from the EU Joint Research Centre [17] [18]; cottonseed oil is not included in any of the analysed pathways. Besides ensuring that biofuels offer consistent GHG direct emission reductions compared to their fossil fuel counterparts, EU regulations aim at preventing indirect land use change (ILUC). This phenomenon was first documented in a 2008 study, and its effects, although subject to high uncertainties, have been proved to sometimes reverse the benefits of biofuels [19]. RED II introduced for the first time in EU biofuel policies the phase-out of feedstocks that are assessed to induce high indirect land use change (ILUC) effects, which translates into a complete elimination of palm oil as biofuel feedstock by 2030. The latter measure is relevant especially for countries with a current high reliance on palm oil, namely Spain, France, the Netherlands and Italy [6]. This reliance on palm oil is particularly marked in the production of RD: in Italian biorefineries, this percentage is currently in the proximity of 80% [1].

Cottonseed oil may be adopted as an alternative feedstock to palm oil, especially in the contexts most affected by these regulations. The majority of cottonseed is currently crushed and processed into its sub-products, most relevantly cottonseed meal and oil; the latter is then treated in order to become suitable for human consumption: otherwise, it would be toxic. As of today, the global share of cottonseed that stays uncrushed is assessed at around 20% of global production, or 8.5 Mt of seeds [20]. In the most favourable contexts, e.g. in Israel, such cotton wholeseed may be sold as cattle feed (ruminants are the only animals capable of coping with cottonseed toxicity), but it may also well become waste: this is especially the case for countries that do not feature a modern cotton industry with a solid supply chain, as for many cotton-producing African countries [21]. If the upgrade of such uncrushed seed were completely devoted to RD production, it would globally yield 1300 kton of oil, which would cover about 50% of EU RD production as of 2017 [6]; this corresponds to covering about 42% of global RD production [22]. Cottonseed oil has not been subject to the extended life-cycle analysis that most other biofuel feedstocks have undergone: to the author's knowledge, there is only one peer-reviewed LCA study dealing with cottonseed-oil biofuel production in rural Brazil [2]; this study investigates cottonseed-oil biodiesel (FAME) production with a batch reactor after oil extraction through simple press, a technology representing a rural context in Brazil. Only one LCA study of cottonseed-oil considering solvent extraction was found, similarly dealing with biodiesel production and a rural context in India, but such study did not undergo any peer review [3]. Existing literature on cotton mostly deals with the life cycle of cotton lint or cotton fabric, which are of priority for the clothing industry [23] [24] [25] [26].

This study investigates the GHG intensity of cottonseed-oil RD through a LCA, in a context where cottonseed is fed to cattle in the form of wholeseed. Two objectives are pursued: (I) define cottonseed oil in the framework of EU biofuel regulations and quantitatively assess the GWI of cottonseed-oil RD, in order to check its compliance with said regulations; (II) assess the GWI of this biofuel in comparison with other feedstocks, in the framework of the most widespread guidelines in matter of life-cycle assessment, i.e. ISO guidelines. The core of this research consists of a LCA: it replicates the features of the product system leading to RD fuel from cottonseed and accounts for all relevant life-cycle GHG emissions. The model has been developed with the support of GaBi, a LCA software. Its main functionality is the automatic calculation of the results once the model is complete; in addition, it provides a database with good coverage of some processes of common use, most notably transportation and energy conversion processes. Modelling a product system in GaBi makes use of three instances called flows, processes and plans; while flows connect processes with each other and with the environment, plans are a means to group processes in a hierarchical and rational way, providing flexibility to the modeller. The LCA developed in the present work is a case study involving Israel for cottonseed cultivation and Italy for biorefining and distribution. The Middle Eastern country is home to a small but advanced cotton industry, where all producers are gathered around a single consortium. A distinct model has been developed for each of the two study goals; the EUC model abides EU guidelines and is thus fit for assessing regulatory compliance; the IS model, on the other hand, abides ISO guidelines and represents the system from a more holistic viewpoint, allowing for a punctual comparison of cottonseed oil with other feedstocks. Note that the nature of the case study does not allow for indiscriminate expansion of its results: while the IS model examines the range of carbon footprint and the trade-offs of cottonseed-oil RD, also in comparison with other feedstocks, any other context requires a case-specific analysis to get to accurate and unambiguous conclusions about its carbon efficiency. The product systems of each model and their boundaries are here depicted in figure F1.

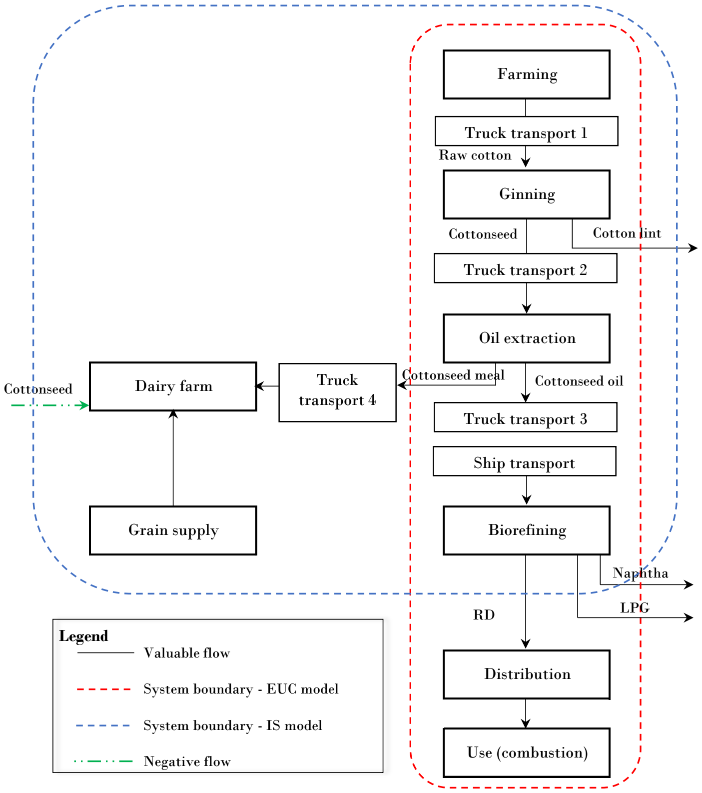

Figure F1: Representation of the two models, highlighting their system boundaries: EUC (red dashes) and IS (blue dashes). Waste and emissions are omitted.

The models differ also in their treatment of co-products: the EUC model ignores any GWI contribution upstream of cottonseed at the gin output, thanks to its regulatory definition as "agricultural residue"; thereafter, it invariably features energy allocation, both at the extraction plant and at the biorefinery outputs. The IS model features system expansion to account for meal use as cattle feed; since this study deals with a diversion of wholeseed to produce biofuels, meal can only partially compensate for this cottonseed, in terms of nutritional value; consequently, a feed supply is included in the IS model. Since the cottonseed flow exiting *Oil extraction* (positive) and the flow entering *Dairy farm* (negative) counterbalance, this model is able to neglect any GWI contribution upstream of cottonseed. Note that this applies as long as the prior use of cottonseed is as cattle feed, as in this case study; otherwise, it would depend on modeller's assumptions on co- and by-product treatment. Throughout model development, the limited data availability required assumptions to adapt available data to cottonseed. Key assumptions are coherent with the goal and scope of the study, which investigates a GWI range rather than a punctual value. A conservative approach is always kept as guiding principle of the study.

The first processes featured in the studied life cycle are cultivation and ginning. For the different reasons mentioned above - regulatory in EUC and model-linked in IS - neither requires their explicit modelling. The oil extraction process was modelled after a solvent extraction facility, which are present in Israel for soybean, sunflowerseed and other oil sources; for such plants, no references for cottonseed are available in the literature: after an analysis of various oil extraction plant, soybean was taken as reference, since it shares the same plant layout as cottonseed. Other oil sources were included in the analysis, for a total of 5 sources.An oil yield was first assumed based on literature data. Observation of different solvent extraction plants shows little variation in oil yield, with values in excess of 95% and approaching unity. Assuming that an existing oil extraction facility in Israel would not be equipped for seed delintering (cottonseed is in fact covered with thin hairs of cellulose, which may decrease oil yield if not removed), a conservative value of 95% was chosen as baseline, with a 90-99% range of variation. In the model, oil yield and oil leakage into cottonseed meal are then parametrised as function of oil yield and seed oil content; this last value is subject to remarkable variability: after observations that place most values in the range of 16 to 20% [98] [99] [100] [101] (although with reports of contents as high as 25% and as low as 10% [103] [104] [105]), it was assumed at 18%. These are both relevant assumption, tested in the results with parameter variation. For utilities consumption, given the lack of data on cottonseed, a functional relation linking these to seed characteristics was investigated. After data analysis, steam consumption is assumed to be proportional to as-received feed; data show in fact an average of 0.61 $^{MJ}/_{kg}$ of seed, with a standard deviation of 10% (versus 1.85±40% $^{MJ}/_{kg}$ per unit of net oil output). Electricity consumption is assumed instead to be proportional to the net oil output: the data are centred on 0.12 $^{kWh}/_{kg}$ of oil, with a standard deviation of 17%; per unit of as-receiver feed, the variability is higher (0.043±23% $^{kWh}/_{kg}$). The baseline for this study features the values for soybean as in (JRC,2017) [18]: for steam, 0.58 $^{MJ}/_{kg}$ of feed; for electricity, 0.15 $^{kWh}/_{kg}$ of oil. To this last value, an extra consumption caused by the harder hull of cottonseed compared to soybean was accounted for by disaggregating this process in the soybean plant and adjusting its consumption to the processable mass flow rate of a commercial dehulling machine (which is higher for soybean and lower for cottonseed, in a ratio of around 40% on average). This resulted in +20% consumption, i.e. 0.182 $^{kWh}/_{kg}$ of oil.

In the modelling of the biorefining plant, the core of which are the hydrogenation and the isomerisation reactors, a distinct approach was applied to utilities and the chemical reactor itself. With regards to utilities consumption, it was possible to rely on models of an Ecofining plant from the literature [16] [137]. The two sources model HVO production without considering isomerisation; notwithstanding, the second of these sources reports an additional consumption of +10 to +30% when producing jet fuel: this allows to assess additional consumption certainly below this threshold. The reactor section is modelled after the two reactions of hydrodeoxygenation (HDO) and decarboxylation (DCO2). After relating the oil feed to the fatty acid composition of cottonseed and by assuming complete conversion (a fact supported by the literature on these reactions [10]), oil and H_2 feeds are linked to products via two parameters: HDO selectivity S_{HDO} and naphtha yield Y_{naph}. Naphtha formation is linked with cracking reactions in the isomerisation reactor, which also generate an extra yield of light products Y_{C1-C4}. This unwanted side-reaction is impossible to model directly without exact thermodynamic and kinetic data: the modelling of the relation between naphtha and light product formation was thus based on available data, which are limited to soybean and palm oil at two different cloud points T_{cloud} [10, p.125]; these are reported in table T1.

Table T1: yields of palm and soybean oil in Ecofining, depending on the final T_{cloud} [10].

	Palm oil		**Soybean oil**	
Cloud point	0°C	-8°C	0°C	-8°C
CO_2	5.4	5.4	5.4	5.4
H_2O	8.3	8.3	8.3	8.3
C_1-C_4	5.9	6.9	6.6	7.0
Naphtha	2.9	9.5	7.7	9.6
Diesel	80.3	75.5	75.5	73

Two observations can relate these oils to cottonseed oil: (I) longer n-paraffins naturally have higher T_{cloud}, which implies that, *ceteris paribus*, they require more isomerisation fixing a desired T_{cloud} [10]; (II) longer n-paraffins are more reactive to cracking, thus implying a higher abundance of light products at fixed T_{cloud} [140]. The assumption is thus that the average carbon-chain length of an oil is a good predictor of cracking, i.e. light products and naphtha yields. Since cottonseed is roughly midway between palm and soybean oil in terms of average length, its cracking products will yield in-between what these two feedstocks yield, ceteris paribus; the result is that the isomerisation reactor yields around 0.21 kg of light products per kg of naphtha. Such modelling of the reactor allowed to also account for the H_2 consumed in the isomerisation reactor. The tuning of S_{HDO} and Y_{naph}, instead, was not based on literature data. By comparing literature with the reactor chemical model, these were found to overestimate yields in ranges of S_{HDO} close to unity and Y_{naph} close to null [16] [18] [137] [141], an ideal situation which does not match industrial reality. Studies also frequently neglect the feed fatty acid composition, which is an important driver of final yields. A consultation with an expert at the Italian operator of RD plants allowed to place actual naphtha yield between 7 and 13%, with proportionally high light products formation [142]; two pieces of information supported the determination of S_{HDO} and Y_{naph}: (I) the information on CO_2 and H_2O yields as from table T1 (which are all identical), where CO_2 can be considered a marker of DCO2 occurrence and H_2O of HDO; and (II) the fact, cited in the literature [10], that the reactor uses a mixed catalyst which is reactive to both HDO and DCO2. Based on such information, S_{HDO} was set at 65% and Y_{naph} at 8.5 wt%: the resulting diesel yield is 74%, while CO_2 and H_2O yields match industrial data. This set-up is conservative, since it corresponds to a low T_{cloud}, reflecting RD production in European winter [13].

The modelling of grain supply for the IS model proceeded from the modelling of three distinct grains: maize, wheat and barley; grains were chosen because they are the typical export-grade caloric feed: since Israel imports nearly the entirety of its animal feeds, they were modelled as imported from the US or the EU, which are the two main exporters to Israel, following Ukraine [106]; this last country could not be modelled since agricultural inventories from Ukraine are not available. Three different feed supply schemes have been modelled: the baseline scheme (BL) features maize and wheat from the US; the other schemes feature barley from Denmark (BDK) and wheat from the EU (WEU) respectively. The BL scheme estimates feed requirements of maize and wheat with a set of two equations, balancing calories (in the form of "net energy for maintenance in cattle") and available proteins respectively. In the other two schemes, a single equation balancing energy was sufficient: in the modelled range for oil extraction yield (90-99%) and seed oil content (16-22%), in fact, the protein constraint is always met (with a small excess of 1 to 5%). Maize, barley and wheat were modelled after consulting eight different sources; they were first uniformed applying a cut-off to inputs in seedling material, pesticides and herbicides, and by evening system boundaries to the farm gate. Sources were found to agree on their main inputs, i.e. fertilisers and fuels [18] [122] [125] [126]. Since US maize is primarily cultivated in the Corn Belt, it was decided to rely on data from this area: of the two datasets from this region, the most conservative was chosen [121]. System boundaries from this source were extended to account for on-farm drying and storage, using European data on the subject [18]. For the BDK scheme, a dataset from Denmark is used [122]. For the WEU scheme, the dataset used is the one in (JRC,2017). This dataset features wheat targeted specifically at ethanol production, i.e. low-grade wheat and not durum or high-quality wheat of higher environmental impact: it is thus suitable for reflecting feed-grade wheat as well. While K and P fertilisers, and lime relied on available datasets from the literature, the importance of N fertilisers in agricultural systems required for a more accurate approach. Fertiliser GWI was thus modelled separately for the US and the EU, keeping into account the difference in supply and imports, and the difference in emission intensity of the production process based on the geographical origin; Emission factors were available for nine regional aggregates [30] [131], while import data were available from official statistics [18] [32]. While keeping track of the emission estimates for all eight datasets, the baseline N_2O emission factors utilised in this research have been uniformed by following IPCC tier 1 methodology [135]. A tier 2 method, based on the *Stehfest and Bouwman* model, has been applied as alternative for the BDK scheme, where accurate soil and climate information were available; input data to the model were sourced from both the LCA study and an online GIS database, known as GNOC [122] [136].

Transport processes were modelled according to the following set of parameters: distance, utilisation ratio (also known as load factor), share of road categories, required sulphur content and share of biogenic C in fuel; among these, the most impacting for GHG emissions are distance and utilisation ratio, estimated using literature data. Gas and electricity supply processes were available in the database, with the exception of data on Israel. Here, European technology was assumed to represent gas supply effectively, whereas the electricity grid mix was modelled after electricity production data from the IEA [143].

ILUC effects, caused by the increase in grain demand, were modelled separately from other emissions; their estimation is based on an analysis of the general equilibrium models GTAP and MIRAGE, which are more suitable to represent a global phenomenon such as increase in grain imports in Israel. After an analysis of both models, and after documenting some scepticism over the last GTAP estimates in the literature [148] [149], it was decided to rely on the latest MIRAGE estimates available [93]; This same study was the one used for reference values of ILUC emissions in EU legislation amending the Renewable Energy Directive (RED), published in 2015 [150]. In the present study, the underlying assumption in grains ILUC emissions estimation is that their effect can be related to grains used in the bioethanol industry, for which emission factors are available in $^{g_{CO2eq}}/_{MJ}$ of ethanol. Such emission factors were related to RD by converting them per unit mass - of grain or oilseed - upstream of the biorefinery: to this purpose, conversion efficiencies and fuel properties had to be assumed based on standard practice; the same was done for cottonseed and the other feedstocks for which a comparison is to be made (palm and rapeseed) [93] [18] [7]. In case of oil biorefining, the emissions per unit of feed had then to be allocated to the specific hydrogenated product of interest, namely Renewable Diesel (RD), by means of energy allocation. The final emission factor then becomes 51.8 $^{g_{CO2eq}}/_{MJ_{RD}}$ for both palm and rapeseed. For grains, the lack of co-products in the use of wholegrain must be accounted for, as opposed to the grain-to-ethanol pathway, which generates considerable amounts of distillers' dried grains. The effects of such co-products was removed based on an alternative scenario of the same publication on MIRAGE estimates, which registers an increase of +46% for maize and of +28% for wheat ILUC factor [93, p.32].

The impact category investigated in this study is *impact on climate change*, through the use of the category indicator *radiative forcing*, which is the physical phenomenon linking GHG emissions with climate change. The characterisation factor utilised is *Global Warming Potential (GWP) 100 years.*, which classifies CH_4 and N_2O as being respectively 25 times and 298 times more potent GHGs than CO_2. The EUC model baseline results in a Global Warming Intensity (GWI) of 15.7 $^{g_{CO2eq}}/_{MJ}$ of renewable diesel, while the IS model baseline stands at 83.7 $^{g_{CO2eq}}/_{MJ}$; in the latter model, feed supply emissions are found to represent about 70% of the total GWI, which makes up for most of the difference. The range of variation - linked especially to differences in seed oil content and oil yield, but also to specific energy consumptions and seed moisture - was assessed at about ±5% for the EUC model, whereas the IS model features a variation of about ±15% (shown as whiskers in figure F2, for feed schemes BL and WEU). This remarkable difference is caused by the aforementioned parameters, which all contribute synergistically to a decrease or increase in impact of the extraction plant; while energy allocation downstream of this unit in the EUC model allows to distribute this change among the co-products of oil (30%) and meal (70%), in the IS model all the burden remains within the product system. Moreover, the system expansion applied in the IS model implies that a change in nutritional properties or quantities of meal impacts on feed supplement requirements: both a lower seed oil content and a lower oil yield concur to higher feed supplement requirements, which is the most relevant and sensitive source of emissions.

At a GWI of 15.7 $^{g_{CO2eq}}/_{MJ}$, the GHG emissions reduction of the EUC model surpasses 80%: cottonseed-oil RD thus seems to comply with EU regulations, which ask for a reduction of 60% (for Italian plants). Furthermore, the contained range of variation seems to make this outcome solid and reasonably applicable to other contexts similar to our case study without important turnovers, making cottonseed-oil HVO a feasible feedstock option for European biorefiners.

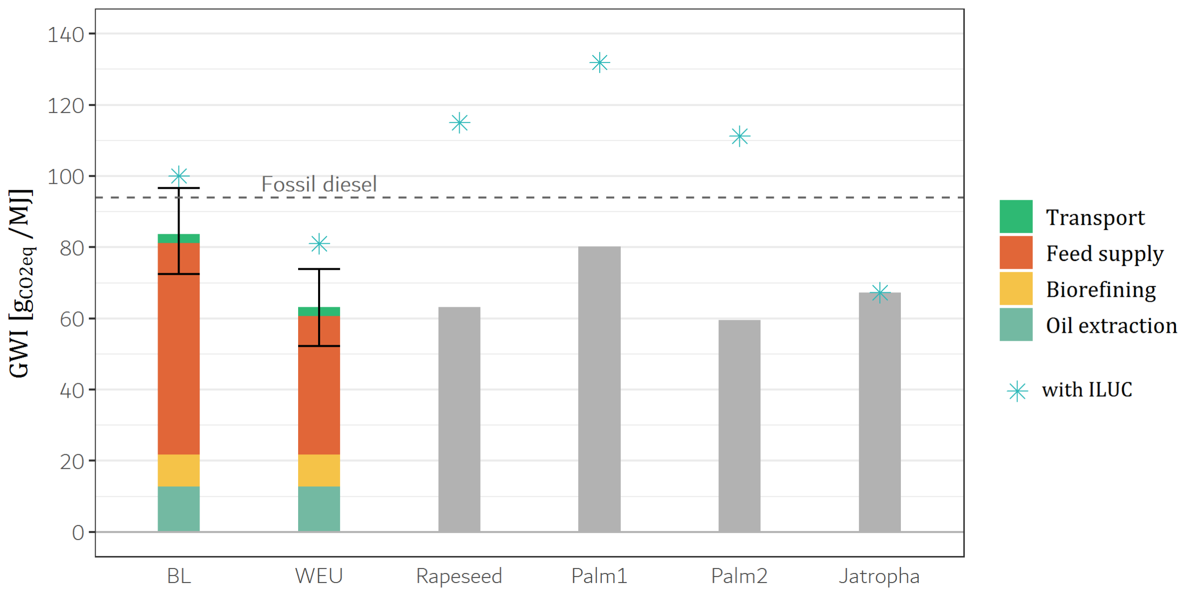

Figure F2: IS model results for BL and WEU schemes, compared to other RD feedstocks (Palm1: palm without CH_4 capture; Palm2: palm with CH_4 capture).

In the IS model, the most relevant share of GHG emissions originates in the supply chain of grain feeds. 20 $^{g_{CO2eq}}/_{MJ}$ separate the BL from the WEU scenario (with scenario BDK in-between these two), as figure F2 shows. In terms of comparative analysis, figure F2 points out how changing feed supply could make up for a lower GWI than rapeseed and jatropha biofuels, in terms of direct emissions. The different carbon intensity of grain production is the first cause for this difference: US wheat - by far the prevalent grain in IS BL feed supply scheme - is the most emission-intensive cereal considered in the analysis, given its higher-than-average fuel consumption and fertiliser input, and low grain yield. A second source of divergence lies in the emissions of grain transport to Israel (which are shown as part of "feed supply" in figure F2). Under the BL scheme, in fact, the supply of American grains increases the final GWI by about 16 $^{g_{CO2eq}}/_{MJ}$, constituting about 20%

of such GWI, while WEU features about 10 $^{g_{CO2eq}}/_{MJ}$ of such emissions. These values also stress the importance of short feed supply chains for reducing the biofuel carbon footprint.

The parameters of seed oil content and oil extraction yield are both relevant in determining the final fuel GWI. Ceteris paribus, an extra 1% of oil in dry seed changes the GWI by around 5%: higher oil prompts less feed requirements since, ceteris paribus, the amount of seed for producing a unit of fuel decreases. Differently from oil content, oil yield brings about two competing phenomena: (I) the necessary seed for a unit fuel decreases; (II) the caloric properties of a unit mass of meal decrease. Just like with oil content, the first effect is beneficial for GWI; the second effect alone, instead, would bring to higher feed requirements. Net of these two contrasting effects, feed requirements are observed to decrease with oil yield, so that the effect of yield on GWI is beneficial. Here, the delta is less impacting: an extra 1% of oil yield roughly causes a decrease of 0.6% in GWI. As for oil yield, two competing effects exist also for reactor selectivity to HDO, respectively given by a change in hydrogen demand and diesel yield; in our case, the trade-off is in favour of DCO2: the reduction in H_2 supply, in fact, more than outsets the increase in emissions upstream of the biorefinery - linked in particular to increased feed supplement requirements and increased utilities consumption at the extraction plant. The effect of naphtha yield is much less marked: changing naphtha yield from baseline to our assumed maximum, i.e. from 7 to 13%, changes the GWI by 0.1%. This is due to the fact that naphtha yield only changes the final proportions of useful products (LPG, naphtha and diesel), which are then assigned with GHG emissions through energy allocation. N_2O emission factors have also been tested: besides the tier 1 method used as baseline, declared emission factors as well as the tier 2 emission factor for the BDK scheme have been compared. In general, declared emissions and *tier 2* emission tend to lead to a result that is close to the tier 1 baseline, with a difference between 1 and 5 $^{g_{CO2eq}}/_{MJ}$ on the GWI. By applying to BL and WEU feed supplies the aforementioned ILUC factors, ILUC emissions of respectively 16.4 and 17.8 $^{g_{CO2eq}}/_{MJ}$ arise, against an increment of about 50 $^{g_{CO2eq}}/_{MJ}$ for other first-generation oil feedstocks. The large uncertainty of ILUC estimates does not allow for solid and precise conclusions; nonetheless, starting from the consolidated observation for which cereals have a lower ILUC effect than oilseeds and at the light of these results, it is possible to conclude that cottonseed-oil RD would lead to consistently less ILUC emissions than the common first-generation feedstocks. Finally, the consequences of using a simple press to extract the oil instead of a solvent extraction plant have been tested, based on data for cottonseed [2]. Such replacement spurs a severe decrease in oil yield (from 95 to 56%): a 25% higher GWI is registered for the BL scheme.

In conclusion, the most relevant outcome is that cottonseed-oil RD is likely to be compliant with EU biofuels regulations, meaning that its use can effectively concur to the attainment of national and EU-wide renewable target in the transport sector; given the contained range of variation of its GWI, such compliance seems certain also in contexts different from the present case study. The IS model indicates GWIs lower than fossil diesel, where feed-supply carbon efficiency is a key determinant of the fuel GHG performance; an efficient grain production and a short feed supply chain seems to be able to deliver a biofuel with lower direct GWI than palm and rapeseed oil, in the range of 50 to 60

$^{g_{CO2eq}}/_{MJ}$. Furthermore, the inclusion of ILUC effects favours the analysed cottonseed-oil RD over competing feedstocks; together with jatropha, of null ILUC effects, the fuel under feed supply scheme WEU keeps below fossil diesel in terms of GWI, as opposed to palm oil or rapeseed oil. The potential carbon efficiency of this fuel pathway thus seems to be promising, with the right feed supply chain. It finally emerges that the methodology used in sustainability assessments – here in terms of climate change – influences the outcomes. EU methodology, while providing an unambiguous set of rules for GHG savings assessment, was not able to include an important portion of the impacts, with a substantial distortion of the results. Regulators must be aware of the possible flaws that a rigid methodology may bring with.

In the medium term, cottonseed may contribute to fill the gap caused by the phase-out of palm oil. In this respect, Africa appears to be a promising region with yet much unexploited potential of seed being wasted. Applying simple press, the technology that would most likely be implemented in rural contexts, has been shown to increase the emissions intensity of the fuel, with an even greater dependence on the feed supply chain that could make it more carbon-intensive than fossil diesel. When designing this process in such a context, effective local value creation and avoided distortion of local markets are imperative issues to be taken into account, together with a carbon-efficient feed supply if wholeseed is used as cattle feed. Further research on the topic may focus on specific contexts, to assess the sustainability of local biofuel supply chain and to verify its applicability. Other interesting outcomes may come from comparing a large sample of feed supplements in its impact on the fuel GWI, depending on their different origins and agricultural practices. Studies covering impact dimensions other than climate change could give additional evidence on the effective sustainability of this biofuel.

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Introduction

Climate change has been recognised as one of the major challenges of our age. The sheer magnitude of this phenomenon is especially evident when contrasting the current levels of average temperature increase and CO_2 atmospheric concentration with the IPCC goal of "well below 2°C above pre-industrial level" temperature increase by the end of the Century. While the magnitude of the influence exerted by anthropogenic GHG emissions in alimenting this phenomenon is yet to be quantified, most countries have put decarbonisation in their agendas. In this context, the role of policy-makers is prominent, especially in the framework of economic activities: here, policies are of vital importance to confront global emissions, which are a form of externality - even labelled as "the greatest market failure in history" by the economist Nicholas Stern [35].

The transport sector represents a primary target for policy-makers, well aware that it alone represents about a quarter of global GHG emissions and that it is still lagging behind other energy sectors: for instance, driven by modern renewable energy technologies, renewable electricity generation has experienced an exponential growth in the last decade, starting from a global share of about 3% in 2010 and reaching almost 10% in 2018 [36]; on the other hand, the same timespan saw the share of biofuels in transport energy consumption rise of just one percentage point, from 2% to 3% [4] [5]. The role of policies in the transport sector is fundamental: the importance of regulations, in fact, is such that biofuel production volumes in Europe are almost exclusively explained by the targets set by biofuel mandates, with tax incentives and oil barrel price exerting only secondary effects [6]. In the European Union, these regulations take the form of directives and implementing acts that set common guidelines and binding targets: the most recent of these directives sets a Union target of 14% by 2030 for what concerns the share of renewable energy used to fuel the transport sector. The previous directive, expiring in 2020, set a similar target of 10%; in 2017; the most recent European Commission (EC) report on the matter expects it to be reached by 2020 at EU level, although some Member States (MS)s still appear far from this 10% blending target [37].

The new target of 14% by 2030 is expected to be met primarily via an increase in the share of biofuels [8] [9]. Policies must thus ensure that such additional reliance on biofuels will not come about to the detriment of natural ecosystems, nor of food and feed markets. Recent EU directives entail a number of measures tackling this aspect, the most relevant of which is the progressive phase-out of palm oil as feedstock. This measure was necessary to limit the use of what is seen as a high Indirect Land Use Change (ILUC) feedstock - meaning a fuel that indirectly causes the displacement of lands, especially with high carbon stocks, such as the tropical forests and peatlands of South-East Asia where most oil palm cultivation occurs. This replacement impoverishes the land carbon stock, with a net transfer of GHG from the ground level into the atmosphere. As of 2018, about 19% of European biodiesel and RD production relied on palm oil, the main importers being Spain, France, the Netherlands and Italy [6]. This reliance on palm oil is particularly marked in the production of RD: in some cases, this percentage can get as high as 80% [1]. It is thus imperative for producers in the aforementioned countries to find new viable feedstocks and differentiate their supply.

This research focuses on the study of cottonseed oil as feedstock in a biorefinery devoted to the production of renewable diesel. The priority, in this sense, is the analysis of cradle-to-gate life-cycle emissions of such biofuel under the framework of a Life Cycle Assessment (LCA). There is currently a research gap in the field of biofuels from cottonseed oil and their related environmental impacts. To the author's knowledge, there is only one peer-reviewed LCA study dealing with cottonseed-oil biofuel production in rural Brazil [2]; in this study, the fuel produced is biodiesel and the oil is extracted via simple seed crushing, two radical differences with respect to the current research. Only one LCA study of cottonseed-oil considering solvent extraction was found, similarly dealing with biodiesel production and a rural context in India, but it did not undergo any peer review [3]. Existing literature on cotton mostly deals with the life cycle of cotton lint or cotton fabric, being addressed primarily to the clothing industry [23] [24] [25] [26].

This research aims at overcoming these issues by developing an original, context-specific analysis that abides by international guidelines. This research is based on a case study of cottonseed from Israel. Thanks to the cooperation of the local cotton industrial consortium - known as Israel Cotton Board (ICB) - it was possible to access first-hand information about their work. Setting the case study in Israel allowed to work in a small context, where all cotton growers in the country refer to the ICB. The Israeli context is then complemented with the Italian context for the biorefining phase, as the Italian biorefinery sector is particularly sensitive to the phase-out of palm oil by 2030.

This research aims primarily at a quantitative assessment of the environmental impact of cottonseed-oil renewable diesel, in order to verify its compliance with the latest EU regulations.[1] Furthermore, the aim is to assess its life cycle developing a model which abides by International Organization for Standardization (ISO) guidelines, which allow for a more punctual comparison of impacts with other feedstocks, in order to assess the environmental quality of this biofuel from a more objective perspective.

These tasks were implemented by means of a Life Cycle Assessment (LCA). Each model

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[1] In this text, the terms RD and Hydrotreated Vegetable Oil (HVO) can be used interchangeably. In the field of biofuels, the latter denomination is somewhat more general, referring to the product of the process of hydrotreatment of vegetable oils, whereas RD is used to identify HVO in the carbon-chain length range of diesel oils, as opposed to the term biodiesel, used to identify the product of transesterification of vegetable oils.

follows different guidelines and is thus suitable for addressing one of the two aforementioned research aims. The already mentioned lack of past research - for cottonseed processing in general and for the specific process of RD production in particular - prompted for the development of approaches allowing to overcome these issues.

This dissertation is constituted by three main chapters. In chapter 2, the policy framework surrounding biofuels will be discussed, with special emphasis on European situation: first, the current situation with regards to biofuels adoption is introduced and explained; thereafter, the regulatory framework in the EU is discussed, with a focus on the most recent policy developments; finally, the implications of such regulations are discussed, introducing the policy driver behind the adoption of cottonseed-oil hydroconverted fuels.

In the first part of chapter 3, the cotton plant will be briefly characterised through its history, its products and its uses, with a special focus on cottonseed and its subproducts. In the second section, biorefining of vegetable oils via hydrogenation will be discussed: the chapter highlights the differences of this with other common biofuels, explains the chemistry behind the biorefining process and illustrates the main features of the specific hydroconversion process layout that will be considered further on in the research.

Chapters 4 and 5, which represent the core of the present research, deal with the LCA of cottonseed-oil renewable diesel. The former chapter starts by presenting the materials used to compute this assessment, the main features thereof, and a simple example of an LCA to explain in brief the main peculiarities of such models. Then, the LCA is carried out, base on a case study that features Israel and Italy as main setting. The results and their discussion are presented in chapter 5. In its final section, this chapter discusses the main conclusions behind the study results, together with observations about the validity and the potency of the results beyond the specific case study treated.

Figure 1.1: An illustration of the main features of *Gossypium Hirsutum*, the most widespread cotton species in use.

Biofuels in the context of the European Union

Section 2.1 opens the chapter by tracing the key points for understanding the theme of biofuels in the EU. In particular, the section serves the following purposes:

- Introduce the topic of biorefineries in the EU.
- Present a brief overview of the main biofuels in use today.
- By focusing on HVO biorefining, present the feedstock partition currently characterising Italian biorefineries. Here, high reliance on palm oil can be observed.

In section 2.2, the reader is introduced to the most important matters touched by EU biofuel policies. The functions and the highlights of this section, thus, are as follows:

- Briefly discuss the evolution of EU regulations in matter of biofuels, and present the performance of EU Member States (MS) in complying with said regulations.
- Present the most recent developments in EU biofuel policies. From the analysis, it emerges in particular that they require a reduction in use of palm oil, up to complete phase-out by 2030. Biofuels consumed in the EU have to comply with a set of rules, and most importantly with thresholds on greenhouse-gas savings.

In section 2.3, which closes the chapter, the following functions are exerted:

- Present a plausible evolution of the sector to comply with regulations. The conclusion is that biofuels will most likely remain the main renewable source in transport.
- Illustrate options to decarbonise different transport sub-sectors. It emerges that some sectors will likely have to rely on drop-in paraffinic biofuels like HVO for their decarbonisation, most notably aviation.

- Present the range of options for HVO producers to adapt to EU regulations. In such framework, it emerges that their position is rather complex, and that cottonseed oil may be an effective addition to their portfolio.

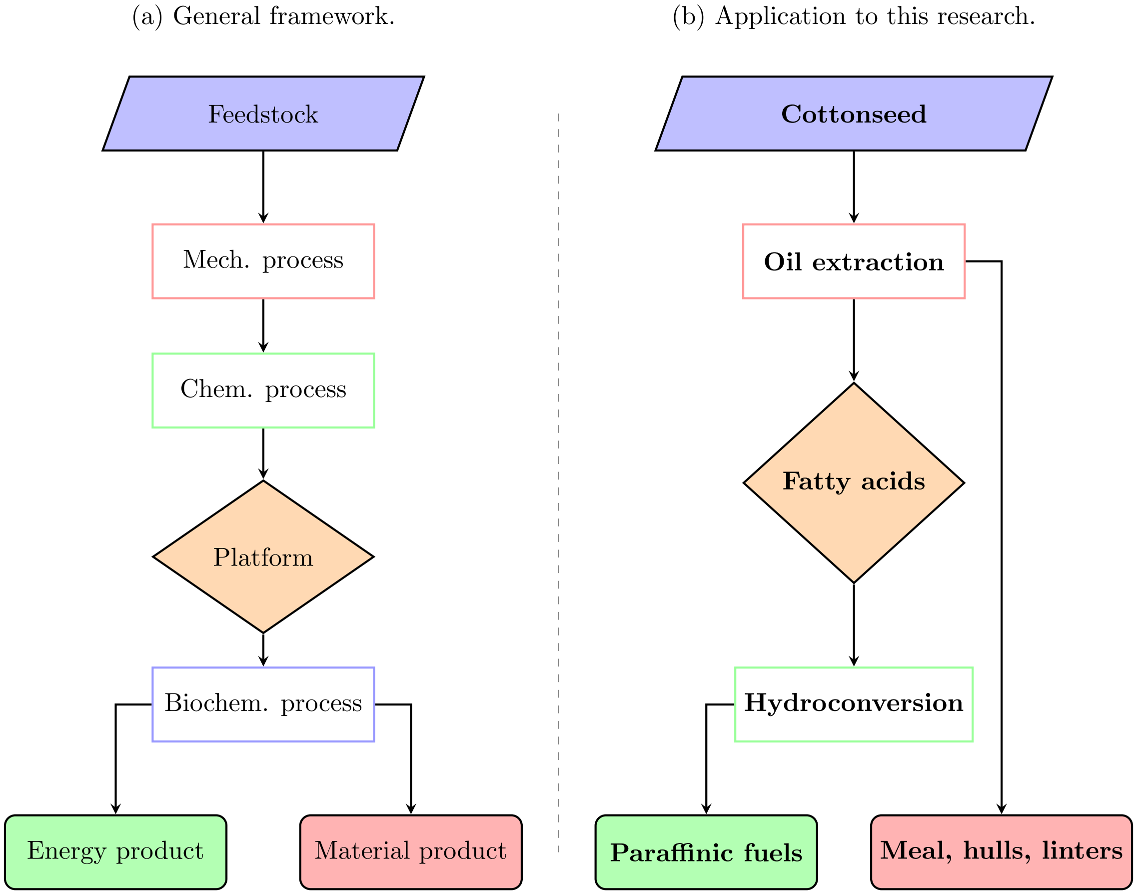
2.1 Overview of the current situation

The main actor in the decarbonisation of the transport sector are biorefineries, in their broadest definition of locations for the *sustainable processing of biomass into a spectrum of marketable products and energy* [38]. In the European Union, the number of biorefineries is estimated to amount to 803 facilities, if we account not only for commercial scale plants, but also for pilot and laboratory-scale plants. The highest concentration is found in the central part of the Union, particularly in Belgium and the Netherlands; 363 of these biorefineries yield liquid biofuels [39].

A biorefinery can be designed to deliver a wide range of products, starting from a variety of different feedstocks and employing an equally diverse set of processes. These three concepts, together with the concept of platform - i.e. any intermediate linking feedstocks to final products - allow to univocally define any biorefinery, from the simplest to the most complex [27]. Figure 2.1 applies this framework (feedstock-platform-process-product) to the context of this dissertation, characterising cottonseed-oil biorefining.

In principle, biorefineries can provide a variety of goods: not only biofuels, but also biochemicals, bioplastics, animal feed - among others. Restricting now our analysis to biofuels for transportation, it is important to briefly illustrate the range of options available today in this field, in order to better understand the drivers at the base of policy reforms in this sector: decarbonisation of transport, in fact, is largely dependent on policy-making, given the substantial cost gap between biofuels and their fossil counterparts; biodiesel consumption in the EU, for instance, is driven almost exclusively by MS mandates and, to a lesser extent, by tax incentives [6].

Feedstocks may originate from agriculture, forestry, households and industries; algae and seaweeds from aquaculture may be considered as a fourth source of feedstocks, but their costs will remain prohibitive without major breakthroughs. A further characterisation is provided by the distinction between dedicated crops and residues. This variety of feedstock options is mirrored in the large number of existing processing options, many of which are still at their early stages of development; this being said, the main actors of current and future liquid biofuel policies can be ascribed in a rather restricted range of options; today, in fact, the most widely deployed technologies are only three: fermentation of sugar and starch crops to ethanol, the already mentioned transesterification of oils and fats to biodiesel, and hydrotreatment to drop-in paraffinic fuels. Other technologies have passed the demonstration stage and are at their early market development; of these, cellulosic ethanol is the most important representative, together with biomethane for transportation from anaerobic digestion [40]. At an earlier stage of development are finally pyrolysis oil and oil from Hydrothermal Liquefaction (HTL), as well as Fischer-Tropsch biofuels.


(a) General framework.

(b) Application to this research.

Figure 2.1: Cottonseed oil biorefining as defined following the biorefinery framework [27].

Focusing on the European market, it can be seen how it has historically favoured biodiesel over bioethanol: the EU is currently the first biodiesel producer in the world, representing more than 40% of the total volume, and the first consumer as well. Besides this, the EU is also an importer of biodiesel. The uplift of anti-dumping duties to Argentina and Indonesia in 2018 caused a consistent increase in imports of Argentinian soybean-oil FAME, and Malaysian and Indonesian palm-oil FAME. This competitive context contributed to a decrease in European production, that amounted to -8% in 2018. Currently, many FAME plants are thus working far from full capacity [6].

In contrast to this trend in FAME production, renewable diesel production has been steadily increasing. From 2016 to 2020, production capacity saw a 43% growth, passing from 2700 million litres to 3870; the most important contributions to this increase was due to new plants in Italy and France entering operation [41]. In particular, Italy has increased its processing capacity from about 360 kton to about 1000 kton, with the start-up of a new plant in Gela, Sicily. Contrary to biodiesel production, renewable diesel is produced in a small number of large-scale facilities. Most of the production originates from stand-alone plants in the Netherlands, Finland, Spain, Italy and Sweden. In some countries, such as Spain and Portugal, co-processing in conventional refineries is also adopted [6].

The dominant feedstock for biodiesel production are energy crops, the most relevant of which is rapeseed, sourced primarily from Germany and France. However, its share in the feedstock mix has been slowly but constantly decreasing, while Used Cooking Oils (UCO) use and palm oil import has been increasing over time. UCO in particular surged from being of marginal importance to being the second most important feedstock: its use in 2000-2003 accounted for 5% of total inputs, whereas today it has reached 22%. However, its use is particularly relevant only in a limited number of countries that managed to establish an efficient supply chain, namely the Netherlands, Germany, Portugal and Austria. Palm oil covers, on the other hand, 19% of total inputs; it is mainly employed in Spain, Italy, France and the Netherlands. Other minor feedstocks include animal fats, sunflower oil and tall oil; cottonseed oil is also employed in minor quantities in Greece [6].

In principle, these same feedstocks can be used for renewable diesel production as well; given the flexibility of the technology, this biofuel tends to rely on different feedstocks depending on location and availability. The three Neste biorefineries operating in Finland and the Netherlands - operated by the largest player in the HVO industry - declare that 83% of their feedstock in 2018 was represented by waste fats and oils, in the form of UCO, Palm Fatty Acid Distillate (PFAD), animal fats and technical corn oil. Another Finnish HVO biorefinery operator, UPM, utilises tall oil, a waste of the wood industry; tall oil is the feedstock of choice also in Sweden, given the relative abundance of this forestry by-product in many Northern countries.

Biorefineries in France, Italy, Spain typically feature a mix of UCO and other waste oils and fats, together with a large portion of imported palm oil. In this respect, the two hydroconversion plants in the Italian context, situated in Gela and Marghera, currently rely on palm oil for 80% of their total feedstock; this is shown in figure 2.2 [1]. The remaining 20% is represented by UCO and other waste oils and fats.

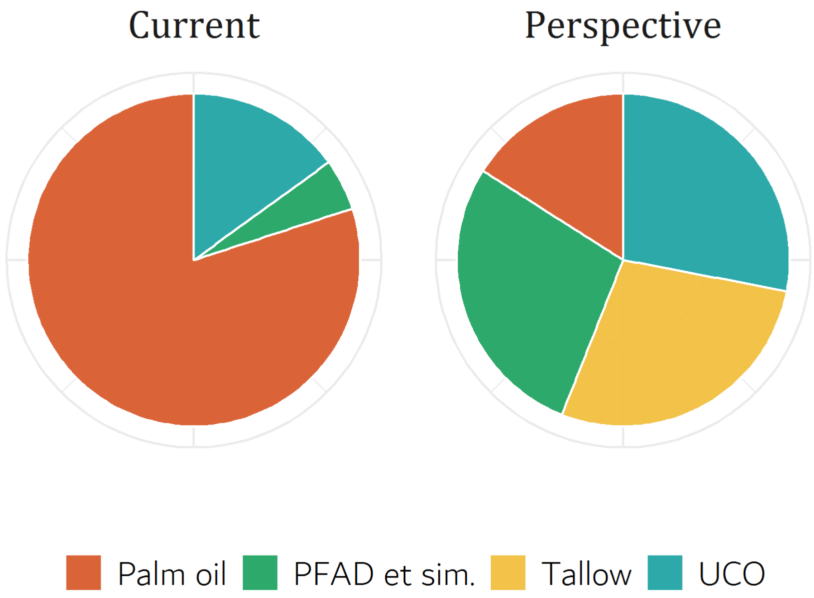


Figure 2.2: Current and perspective feedstock partition in Italian HVO biorefineries [1].

The company has bold plans to strongly increase the shares of waste oils and fats - nearly doubling the UCO component and introducing tallow for as much as 28%: HVO production from animal fats is in fact particularly convenient, given the prevalence of saturated fatty acids in such feedstocks. In any case, their latest projections as shown in figure 2.2 do not yet account for the complete phase-out of palm oil that EU legislation wants to enforce (see section 2.2), as prospects still feature it at 16%. The complete phase-out of palm oil will thus lead to an even more drastic change in feedstock partition.

The bioethanol market has been stagnating until 2016, but thereafter consumption has recovered, with an increase in domestic production as well. This can be explained by higher blending in the gasoline pool, driven by a reduction in the price gap with gasoline and by the will to comply with European targets to 2020 in a timely fashion; In 2019, this trend is expected to continue. 2019 saw the EU setting aside the massive anti-dumping duties that had been imposed on US ethanol since 2013. However, the compliance with EU regulations, most importantly in matter of minimum GHG reduction - explained in the next section - may limit the amount of US bioethanol exports to the EU in the future.

The production of bioethanol is dominated by the use of dedicated cereal crops, such as wheat, maize and barley. Sugar beet is also widely employed. Lignocellulosic bioethanol, which utilises feedstocks such as wood milling residues and dedicated short-rotation forestry, is still on the path of reaching commercial maturity. The first commercial scale second-generation bioethanol plant in the world, which is situated in Crescentino (IT), began operation in 2013, but was forced to shut down in 2017 due to financial problems; it is expected to become operational again in 2020, featuring a capacity of 50 million litres [42]. This value corresponds to the total capacity in Europe in 2019, with plants in Finland and Norway. By 2021, expansion of current plants and new projects in these two countries are expected to add 100 to 150 million litres of capacity; projects are also being planned in Slovakia and Romania [6]. It is nonetheless evident that this sector would need huge commitment to be scaled up to the levels of conventional bioethanol and biodiesel.

Biomethanol, which can be produced starting from biogas, is currently produced in only one facility of 250 million litres capacity in the Netherlands; however, as of 2017, the plant has been working far from full capacity, producing about 75 million litres of methanol [6]. The biomethanol market is much smaller if compared with other biofuels and it exhibits many competitive uses in the chemical sector, but its diffusion - or the diffusion of related products such as bio-dimethylether - as transportation fuel may become a reality in the future, for example for heavy road transport [43].

In 2019, total biofuels blending with fossil fuels in the EU is estimated to have reached 5.7% by energy content, with a bioethanol fraction of 4% and a biodiesel/HVO fraction of 6.4% in the respective pools; food-based feedstock represented the major share of this amount of biofuels consumption, at an estimated 4.6% blending. The advanced, non-food and waste-based fraction constitutes the remaining 1.1%, of which only 0.2% comes from agricultural and forestry residues, while the rest originates from UCO and animal fats [6]. The ambition of the EU is to strongly increase, in the following years, the renewable fraction of its transportation fuels; furthermore, its policy-makers regard GHG reduction and land-use change as the top challenges to confront, together with the fuel-versus-food dilemma: as the next section illustrates, new policies are meant to cause a deep reshaping of the EU biofuels industry in the years to come in order to address these issues.

2.2 Regulatory framework

The European Union is currently under the effect of the so-called European Climate Change Package (ECCP). The first EU directive explicitly covering the whole energy system, known as RED, entered into force in June 2009 and is going to expire on the 31st of December 2020; this directive, which till now has been the regulatory pillar of the Union's energy transition strategy, will be replaced, starting from 2021, by the so-called Renewable Energy Directive II (RED II), whose text was published in December 2018 after two years of intense debate. These directives have the purpose of establishing a common ground for all MSs to then enforce them via national regulations. While establishing a common framework, they leave each MS room with regards to the achievement of the goals outlined. The transposition of the directive into national legislation by each MS is thus an essential step.

The core of the package has been the so-called 20-20-20 targets: 20% cut in GHG emissions, 20% Renewable Energy Sources (RES) in gross final energy consumption, 20% improvement in energy efficiency [44]. For what concerns the transport sector, the main policy instrument of RED are blending targets: the 2020 target is 10% RES by energy content, to be reached by all MSs; this target addresses road and rail transport only, excluding aerial and naval transport.

An important pillar of RED was also the introduction of the GHG savings principle, i.e. the requirement for a minimum GHG saving with respect to the fossil fuel counterpart of any biofuel, under a LCA perspective; this principle is enforced by the introduction of voluntary schemes, which are verification procedures that have to be officially approved by the EU: in short, they represent an alternative to national verification systems for ensuring the compliance of any biofuel to the GHG savings criteria. Other broader environmental sustainability aspects are also covered by RED, such as the refusal of biofuels obtained from land with high biodiversity value.

RED was complemented, in the same year, by the Fuel Quality Directive, laying down a target also for the total GHG emissions from transportation; it also established a 10% blending cap of ethanol in gasoline, and compliance criteria for biofuels by means of additional blending caps.

In 2015, the Commission adopted an amendment to both aforementioned directives called *ILUC directive*. This was done in response to some environmentally harmful effects that the ECCP package had been accidentally causing: in particular, previous regulations lacked any considerations on indirect land-use change effects, which accounts for emissions associated not with direct displacement of high-carbon-stock land to cultivate energy crops, but rather emissions indirectly associated with such cultivation. The rationale is that any diversion of existing agricultural land to the production of first-generation feedstocks translates into a need for more land to cover food demand; depending on many variables, such as geographical distribution of the production, local regulations and global market interactions, this augmented demand may translate in a more or less intense impact in terms of deforestation and land conversion. The effects of land use change on the carbon balance of a biofuel have been proved to sometimes compromise its beneficial effects: in two breakthrough studies from 2008, oil palm plantations replacing tropical forests and peatlands were reported to reach a break-even in the ecosystem carbon balance after about 50 to 400 years of operation [45] [46]. The scientific community first recognised the risks of indirect land use change in 2008, when Searchinger et al. [19] showed that including ILUC effects of corn-based ethanol would double its GWI in a time frame of 30 years, making it more emission-intensive than fossil gasoline.

The ILUC directive was introduced to limit the damage associated with this pressing issue: it capped the production of conventional, food-based biofuels that can be used to meet the 10% target at 7%, and it also required MSs to have a minimum share of 0.5% advanced biofuels. The ILUC directive also introduced a principle that has been the implemented also in the subsequent RED II: the adoption of double-counting. Advanced biofuels, in fact, account for twice their energy value towards complying with the set blending target: for this reason, the 5.7% total blending by energy content that was mentioned at the end of the previous section translates into a 7.3% share for the compliance with the directives, thanks mainly to the harnessing of UCO and other waste fats and oils.

Nonetheless, reminding that the 10% blending target imposed by these regulations is to be reached by each MS, there are appreciable differences with regards to the blending levels so far attained; in figure 2.3, it is possible to observe how some MSs have been clearly underperforming, whereas some others, notably Sweden, place themselves as virtuous outliers [6].

RED II, the new directive covering the decade 2021-2030, is designed to have a much more universal validity than its predecessor: in fact, it covers also some matters related to aviation and shipping transports; it also covers solid biomass, which was ignored in previous legislation.

RED II sets a binding RES target of 32% by 2030 in gross final energy consumption, with a 14% target specific for the transport sector complemented by a clause of possible upward adjustment by 2023; it is important to point out that while the former is a EU-wide target, further developed by setting separate targets for each MS, the latter is common to all MSs.

In the calculation of compliance with this 14% target, blending from food and feed crops is allowed to be maximum one percentage point higher than the MS's consumption thereof in 2020, and not exceeding, in any circumstance, 7%. In addition to this absolute threshold, the directive states that their use cannot exceed one percentage point above 2020 consumption, thus effectively placing a threshold lower than 7% for most MSs.

The directive has a special focus on advanced biofuels. In annex IX, in fact, two

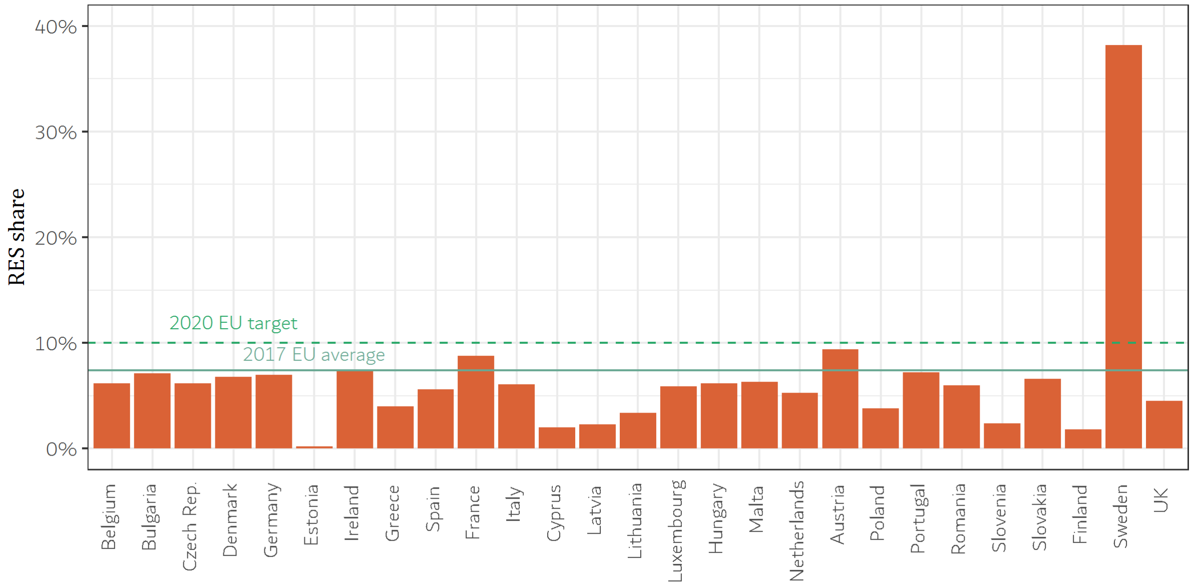


Figure 2.3: Renewable share in the transport sector by EU Member State [6].

different groups of advanced biofuels are identified: *part A* collects mainly forestry and lignocellulosic agricultural waste, whereas *part B* concerns specifically waste fats and oils. Group-specific policies are applied to the two groups: the production of group A biofuels is encouraged, and covered by a series of progressive subtargets that are to lead to a 3.5% share in 2030 - a value that is intended including double-counting, so that the physical blending in energy terms would be half of the aforementioned 3.5%. Used cooking oils and animal fats, included in part B of the annex, benefit from double-counting; however, they are subject to an upper blending threshold of 1.7%, and no binding target is put in place. In the appendices, it is possible to find the complete list of annex IX feedstocks.

This list of advanced feedstocks has been revised from the first RED, dedicating special attention to waste and lignocellulosic materials such as straw and bagasse, and non-food lignocellulosic material in general. The directive allows future legislation to integrate this annex with new feedstocks, while their removal is not allowed.

In RED II, the use of double-counting and similar principles has been extended compared to previous legislation. In the intention of policy-makers, this instrument serves the purpose of encouraging renewable penetration in particularly delicate segments of the transport sector, while at the same time sending a clear market signal for the adoption of advanced biofuels and electromobility. RED II favours in particular advanced biofuels of group A and B with a multiplier of 2, and electromobility, with a multiplier of 4; the complete list of multipliers can be found in the appendices.

RED II also introduces a novel distinction for feedstocks, based on their correlation with ILUC. More specifically, the directive defines high ILUC-risk feedstocks, as "feedstocks for which a significant expansion of the production area into land with high carbon stock is observed" [7]. For these feedstocks, consumption thresholds are blocked at their 2019 levels, with a series of progressive subtargets to limit them further, until total phase-out in 2030. The individuation of high ILUC-risk feedstocks was addressed in a subsequent report from the Commission, published in March 2019; the study concentrates on the most common oil crops, cereals and sugar crops and, after focusing only on the crops with the most cultivated area and relative area increase, it estimates the *share of expansion into rainforest or other land with high carbon stock*. By combining the estimate of net carbon loss from replacement of such land, spread over 20 years, and the average energy yield per unit area, the study assesses the critical threshold nullifying GHG savings at 14% expansion; applying a discount factor of 30% to this result, a value of 10% is found. Considerations on the type of high carbon-stock terrains and on the average yield of the single crops and permanent plantations are also included in the final formula for the estimation of ILUC impact. On the base of a literature review and an assessment based on Geographic Information System (GIS) information, the study concludes that, among the considered feedstocks, only palm oil overcomes the 10% limit, with an estimated 23% share of expansion. This means that, by now, palm oil is the only feedstock intended to be phased out by 2030.

Note that a delegated act following RED II opens for the possibility to certify a specific feedstock source as low-ILUC and avoid the phase-out; this is allowed for feedstocks cultivated in unused land or emanating from crop which benefited from improved agricultural practices. In order to obtain such certification, producers are asked to apply measures leading, in nuce, to: (I) an increase in yield of food and feed crops on land already used for such purposes, that must be proven to be financially attractive only thanks to the validity of additional feedstock under RED II; or (II) to cultivation on unused and abandoned land; or, finally, (III) to cultivation by small holders. By the end of June 2021, these requirements could be amended by the Commission [47]. These conditions make current palm oil production incompatible with this certification.

Finally, RED II establishes a new set of GHG emission savings thresholds for biofuels, depending on the start-up date of the producing plant. The oldest plants, in operation before October 2015, need to comply with a 50% threshold, whereas the newest, operating from January 2021, need to observe 65% GHG savings; plants entering operation in-between these dates must comply with a 60% reduction. This last threshold is the one in force for both Italian HVO biorefineries, and thus the one considered in the present analysis.

In order to assess the compliance of cottonseed-oil HVO with RED II, it must be verified that savings go beyond this 60% threshold, following equation 2.1 as defined in RED II, annex V:

$$SAVINGS = \frac{E_{f(t)} - E_B}{E_{F(t)}} \tag{2.1}$$

In this equation, E_B represents the emissions of the investigated biofuel pathway; the fossil fuel comparator for transport $E_{f(t)}$, on which the savings are benchmarked, is defined at 94 $\mathrm{g}_{\mathrm{CO2eq}}$ per MJ of diesel fuel (LHV); one model will specifically deal with such assessment of E_B for cottonseed-oil renewable diesel, in order to calculate its GHG emissions savings.

RED II also specifies how to account for emissions according to the various life-cycle steps. System boundaries must go from raw materials extraction up to fuel combustion ("well to wheel" perspective), the main contributors to E_B being "cultivation", "processing" and "transport and distribution" stages; according to the methodology, organic carbon ought not to be accounted, both in the form of uptake at the cultivation stage and in the form of release during fuel use. Allocation must be invariably done using energy as criterium, as discussed in the next chapter, and emissions related to the production of machinery and infrastructure ought to be excluded from the analysis. The equation showing all the contributions leading to E_B is reported in the appendices.

This overview of the most recent regulations im matter of biofuels serves as indispensable framework for the considerations of the next section, where the features of the current European biofuels market and its evolution is to be analysed, and the rationale behind the study of cottonseed oil is highlighted. In particular, the crucial aspects to address here are: (I) whether the directive considers cottonseed as a residue or as a product, equally to cotton lint; and (II) whether the directive defines cotton, whose oil is toxic in principle but edible after chemical processing, as a food and feed crop. The latter aspect is relevant to determine whether the aforementioned cap of 7% holds for cottonseed oil as well; the former is relevant for the calculation of its environmental burden, since residues, as defined by the directive, do not share this burden with other products upstream of their production, i.e. the net environmental impact of a residue, according to EU regulations, is null.

The definitions featured in RED II allow to consider cottonseed a residue; in fact, quoting from the said directive: "residue means a substance that is not the end product(s) that a production process directly seeks to produce; it is not a primary aim of the production process and the process has not been deliberately modified to produce it." [7] This is the case for the cotton value chain, which is devoted to the production of cotton and whose production of seed is an indirect consequence of the ginning process. Furthermore, according to RED II, cotton does not belong to the category of food and feed crops. This is in fact defined as "starch-rich crops, sugar crops and oil crops produced on agricultural land as a main crop, excluding residues [...]" [7]. As it will become clearer in the next sections, these two features of cotton are beneficial both for its use and for its compliance with EU regulation in matter of GHG emission reduction.

2.3 Evolution of the market and rationale behind cottonseed-oil renewable diesel

Investigating the future of biofuels is not a straightforward task, given its high sensibility to patterns of technological progress, global market and policy implementation. Key uncertainties, in this sense, originate from the wide range of options to achieve decarbonisation, that include synthetic fuels of non-biological origin and electromobility, together with the peculiarities of each subsector - road transport, maritime transport and aviation.

As already pointed out, policies are of pivotal importance for guiding the decarbonisation of the transport sector. In this respect, the previous section discussed how RED II, the main biofuels policy element in EU legislation, sets three main constraints on biofuels production in the decade 2021-2030: a cap on food-based biofuels, a target for biofuels recognised as "advanced" in annex IX part A, and a second cap on biofuels from Used Cooking Oils and Animal Fats (UCOAF), comprised in part B of the same annex. [1] Figure 2.4 provides a graphical representation of such constraints projected in future EU consumption; they are here deprived of their multipliers, so that values refer to physical quantities.

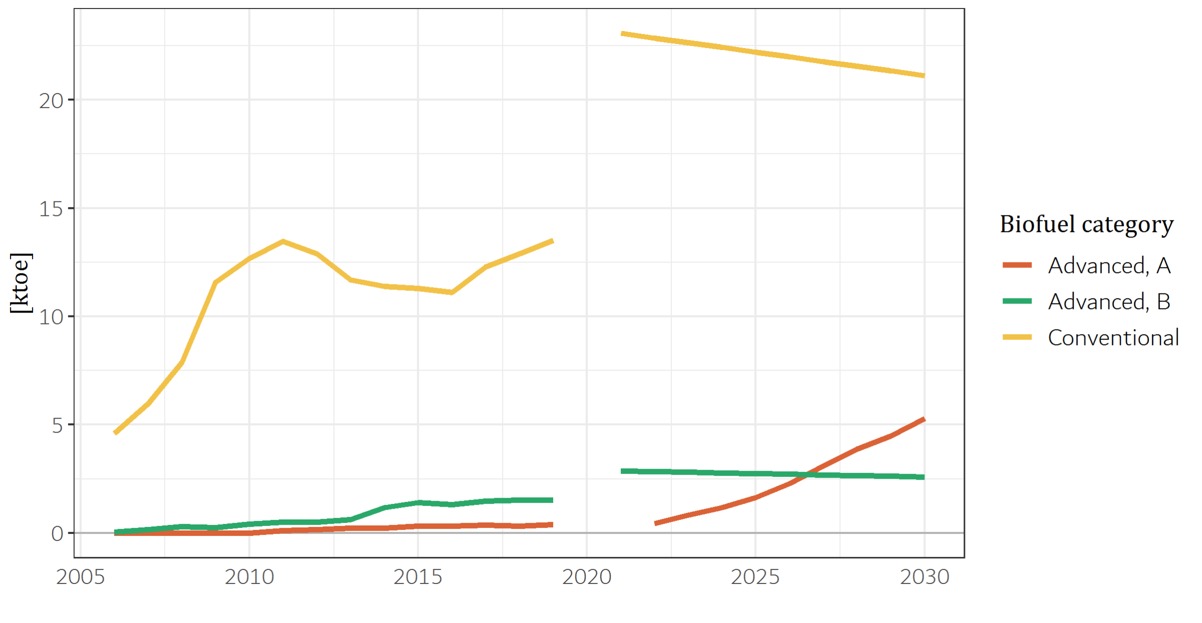


Figure 2.4: RED II thresholds and past consumption in the EU. Quantities of advanced biofuels are reported in physical terms, thus excluding double counting.

As most evident from the cap on food-based biofuels, absolute quantities of both caps reduce with time: the sectorial energy demand is in fact expected to decrease from around 350 Mtoe in 2020 to 320 Mtoe in 2030 [48]. It must be noted that the 7% cap on food-based sources is an absolute maximum, but in many MSs the effective limit will be of one percentage point above their 2020 consumption, i.e. lower than 7%; the policy objective is thus rather to block these feedstocks to current levels.

The graphs reveals that used cooking oil (UCO) and animal fats biofuels (loosely corresponding to *advanced part B* biofuels) have not yet reached saturation as of 2019, on a EU level. In this respect, there is substantial difference across different States on how capillary the supply of these feedstocks to the biofuel industry has been enforced: in 2018, 5 MSs represented 90% of UCO use as biofuel feedstock [6]. In Italy, collected UCO is

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[1] From this point in the text, the term "advanced biofuel" will be used, unless differently specified, to specifically indicate biofuels listed in annex IX, part A of RED II directive.

delivered to biorefineries in the amount of 62 kton per year, or enough to cover about 6% of current capacity; the potential stands at 280 kton: of this, 100 kton would originate from commercial activities, whereas the remaining 180 kton from households; in spite of constant progress in collecting UCO from commercial activities, the latter sector seems much harder to access [49].

Figure 2.4 also shows how substantial the increase in consumption of advanced biofuels type A (i.e. sourced mainly from lignocellulosic feedstocks) should be to comply with regulations, reaching about 5.5 physical Mtoe in 2030. Compared with the current EU capacity of 33 ktoe of second-generation ethanol, the sector - together with other minor advanced biofuels such as biogas-sourced biomethanol - is asked to increase its capacity by more than 160 times in the next decade.

Scientific literature does not offer a univocal forecast for the evolution of the European transportation sector under RED II; however, all studies agree on the importance of liquid biofuels. Figure 2.5, in this respect, shows a scenario for 2030 with regards to alternative energy carriers in EU transportation, as it was presented to the EC in 2016 during the elaboration of RED II: the figure is related to a scenario with 30% RES share in the Union's final gross energy consumption - named EUCO30 in the EC working paper; this scenario is chosen because it is close to what eventually became the Union target through RED II, i.e. 32% by 2030. It reports that 6.2% of 2030 transportation energy carrier consumption is to come from biofuels [8], i.e. 40% of total alternative fuels consumption.

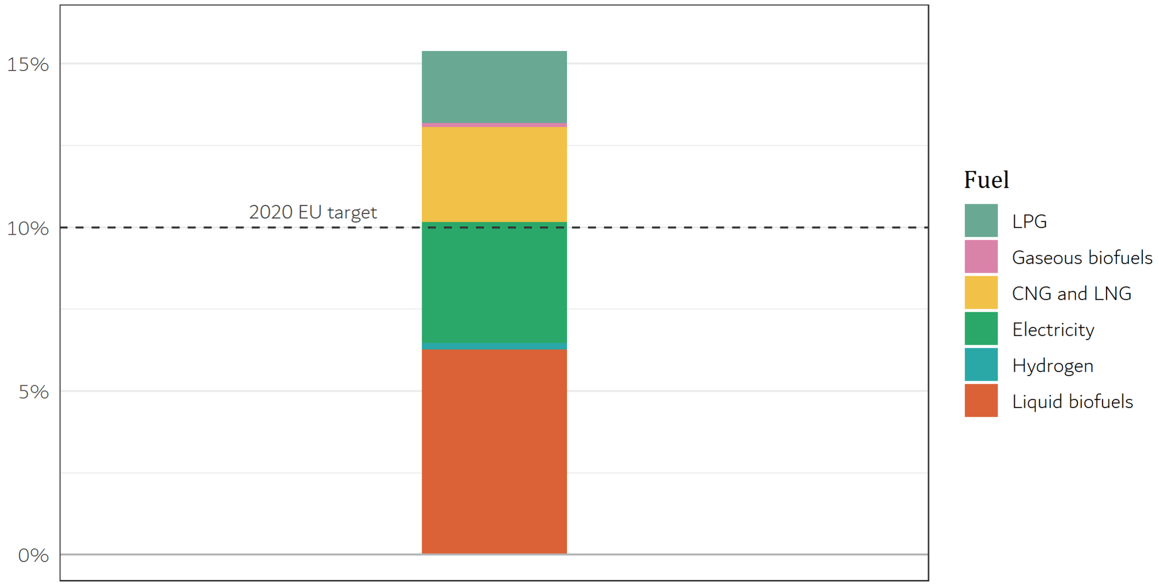

Figure 2.5: Alternative fuels in transportation according to EC scenario EUCO30 [8].

Moreover, this scenario features a solid electricity share in transport energy demand of 3.7%, compared to 1.2% in 2010; the highest increase is foreseen in road transport, with a minor contribution due to further replacement of diesel-powered trains [8]. The scenario projects 5% purely electric vehicles in the EU Light Duty Vehicle (LDV) stock in 2030; pure Internal Combustion Engine (ICE) LDVs are given at 60% of stock in the same year. In terms of market share, the International Energy Agency (IEA) foresees a consistent development of electric LDVs, as they are said to interest about 15% of 2030 sales - which becomes 63% if hybrid technologies are also taken into account [9].

If meeting the target will mean high penetration of electricity for light duty transport, other modes cannot rely much on this pathway to decarbonisation; in heavy road transport, for instance, electrification is forecasted to be much more contained: this situation which calls for alternative solutions, the most immediate being increased blending with biodiesel and HVO; besides the increase in blending rates, this industry must also consider alternative biofuels such as dimethylether and methane, which would enlarge the range of available green feedstocks; obviously, these are not short-term solutions, as they require a redesign of the powertrain [43].

Lately, maritime and aerial transport have both seen the introduction of new regulations, that may result in a stronger demand for biofuels in the coming years. In the maritime industry, the most impacting contribution to global emissions is linked with the transport of goods. Within merchant shipping, a key segment considering that it moves more than 80%of all goods in the world, heavy fuel oil has always been the most consumed fuel. Its cost advantage, however, translates into a poor fuel quality: merchant shipping alone represents 2-3% of global CO_2 and 4-9% of global SOx emissions [50]. The main regulatory agency, in this respect, is the International Maritime Organisation (IMO), operating under the UN. Regulations limiting sulphur content in fuels has long been in place in some coastal waters - known as Sulphur Emission Control Area (SECA) - but from 2020, similar regulations are to be applied to non-SECA areas in Europe as well. The sector will have to pass from 3.5% sulphur to just 0.5%. Many options exist for the industry to adapt: most likely, a first solution will be more severe hydrotreating of fossil fuel oils, in order to eliminate sulphur; this will directly cause an increase in fuel price, together with an increase in hydrogen demand from the refining industry and, consequently, a rise in embodied CO_2 emissions. A second complementary solution is the blending with biofuels, e.g. pyrolysis oil, as they do not contain any sulphur; other options include the refitting of engines and fuel storage systems to employ Liquefied Natural Gas (LNG) as fuel, and the adoption of multifuel engines, that were recently developed to be able to use diesel oil, gas, as well as alcohols in a diesel cycle [51]. Regulations will likely result in an increase in global biofuels demand, be it in the form of bioethanol, biodiesel or HVO; other factors may contribute to an increased biofuels demand as well, such as the possibility to extend limitations also on GHG emissions, both sector-wise and at governmental and local level [50].

Civil aviation is becoming increasingly relevant as a means of transportation. At a global level, sectorial CO_2 emissions grow by more than 4% per year [12]; the same trend is present also within the EU: despite appreciable efficiency improvements - forecasted at +27% in 2030 compared to 2010 levels - the EU projects a growth of 17% in final energy demand in this same timespan [48]. In order to reduce the environmental impact of its aerial transport, the EU launched a programme called *EU Flightpath Initiative*, which aspired to achieve 2 Mt of renewable jet fuel by 2020; the most recent official Eurostat data, however, show that in 2016 the sectorial consumption of biofuels remained at negligible levels, witnessing a substantial failure of this initiative [37]. In order to encourage efficiency improvements and adoption of cleaner fuels, the EU subjected intra-EU flights to its Emission Trading Scheme (ETS) in 2012. Extra-EU flights are instead covered by CORSIA, a programme implemented in 2016 by the International Civil Aviation Organisation (ICAO); the programme's goal is achieving carbon-neutral growth of the aviation sector from 2020 onwards. Until 2023, the programme remains in pilot phase, with non-mandatory adhesion. CORSIA promotes certified renewable jet fuels as a possible solution in the portfolio of operators, together with certified carbon offsetting initiatives. This last solution alone, which is by now the most prominent, has the potential of offsetting the entirety of aviation emissions, although it is argued that, in the absence of additional eligibility restrictions for programmes, CORSIA will not result in significant emission reductions beyond those that would occur without such a scheme [52]; in short, most of the programmes contributing to the carbon footprint reduction of aviation do not really depend on the contributions from the sector. What makes the adoption of biofuels in aviation more complex than in other forms of transport is the restricted range of technical solutions available: differently from maritime and heavy road transport, in fact, the most viable solution seems to be the sole use of drop-in fuels such as HVO jet fuel. The supply of renewable diesel and renewable jet fuel is thus especially important for the decarbonisation of this sector.

It has been pointed out how RED II emphasizes the role of second-generation bioethanol in its path to decarbonisation; currently, its direct use as fuel is currently limited to blending with gasoline, which is a declining market [48]. In the future, its adoption could be fostered by the development of a European market for flex-fuel vehicles and E85 - containing 85% bioethanol; at present, only a small number of MSs have commercialised E85, notably France and Sweden [6].

In maritime, aerial and heavy road transport, the use of lignocellulosic advanced biofuels would require more complex technical expedients than lignocellulosic fermentation; this issue holds especially for the aviation sector, where drop-in biofuels are necessary. In this sense, a 2019 study reviewed all the viable production pathways for advanced jet fuel: hydroconversion to HVO jet fuel; gasification and Fischer-Tropsch process; fermentation and Alcohol-to-Jet-fuel (AtJ) process; Direct Sugars-to-Hydrocarbons (DSHC) via farnesene production; Power-to-Liquid (PtL). The results of the economic analysis are shown in figure 2.6: alternative pathways are from 2 to 8 times more costly than their fossil counterpart, with HVO proving to be substantially more convenient than all other alternatives, especially if sourced from UCO.

Another consideration is the different relevance of feedstock cost: in the case of HVO, it represents more than half of the levelised cost, whereas more premature technologies require a higher degree of capital expenses. The study also highlights levelised cost per tonne of sequestered CO_2, finding that HVO jet fuel sourced from UCO is again the best alternative, followed by Fischer-Tropsch fuels. The conclusion is that the relatively low capital expenditure of hydroconversion, together with the use of residues or waste oils, makes it extremely performing both economically and environmentally [12].

The transport sector will have to undergo radical changes in the coming decade:

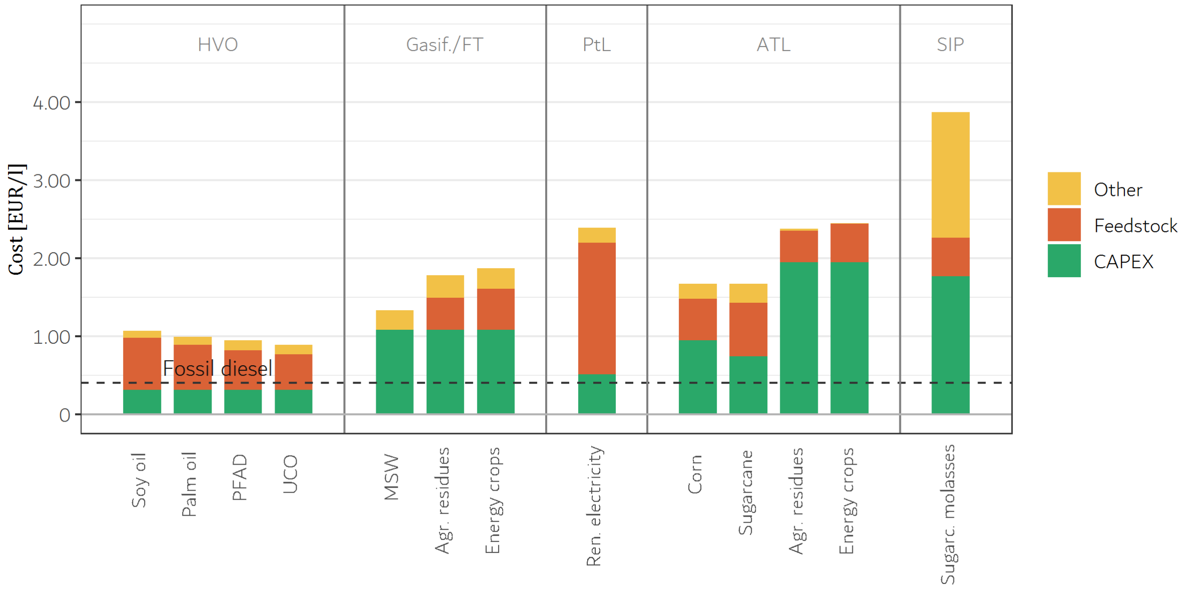

Figure 2.6: Cost breakdown of all the fitting alternative aviation fuels [12].

electrification for LDVs; replacement of diesel with other fuels in heavy transport; very pronounced increase in lignocellulosic ethanol production, with increase in trade among MSs. This last point in particular represents a critical challenge for the sector: the attainment of RED II targets will depend mainly on the efficacy and stability of its policy support with regards to lignocellulosic ethanol, together with a positive business environment. Policies in particular have been recognised as the most impacting obstacle to their development by the industry so far [53]. The capital-intensive nature of advanced biofuel investment, together with the long set-up time of facilities, must be promptly understood and managed by EU policy-makers in the next years, given the proportion of the pursued change.

This being said, most sectors will unlikely be interested by this new industry in the short and medium term, and they will have to rely on drop-in paraffinic biofuels; among such biofuels, HVO represents the cost-optimal solution, provided that it is supplied sustainably. In this sense, many HVO producers are now asked to replace palm oil with more sustainable fuels that are allowed under RED II. This challenge is especially vivid in Italian biorefineries, where palm oil feedstock constitutes 80% of feedstock partition [1].

Italian HVO biorefineries may implement the phase out of palm oil through a number of different options: a first option would be enhancing the production from used cooking oil and animal fats, which is currently below 15% of feedstock partition on national level. RED II features a limit of 1.7% (including double counting) on these feedstocks, a fact that could potentially limit their use - the purpose of such measure being not to obstacle other competing uses. [2] Most importantly, however, even under complete collection of

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[2] In this respect, it is relevant to highlight that the directive leaves room to increase this threshold: "Member States may, where justified, modify that limit, taking into account the availability of feedstock. Any such modification shall be subject to approval by the Commission" [7].

UCO from commercial activities, not more than 10% feedstock share could be covered; household cooking oil, which brings the total national potential above 25% coverage of feedstock needs, poses a challenge that will hardly be met in the short to medium term. As shown in figure 2.2, the operator of Italian HVO biorefineries expects to be able to eventually reach 28% UCO use in its facilities, which would mean complete harnessing of the national UCO potential [1].

A second option is represented by the replacement of palm oil with allowed first-generation feedstock, such as rapeseed oil and soybean oil, within the cap on food-based feedstocks. In this case, Italian operators would likely have to import these goods from other EU and extra-EU countries. This fact may be of concern in future reviews of the regulations, as a possible risk of additional food-based feedstock imports exists, particularly with regards to soybean oil from the Americas; the EC assessed its share of expansion into high carbon-stock lands at 8%, not far from the high ILUC-risk threshold of 10%, but it is not to be excluded that a demand push may induce a higher share of expansion into high carbon-stock land. There are also recent independent studies that already assess a share of expansion in excess of 10% for soybean oil [54].

A third option would be using low-ILUC-risk certified palm oil and similar feedstocks, which would not incur in the ban. As already pointed out, the vast majority of current palm oil production cannot comply with the requirements for such specification.

A fourth option is the use of residues that may be easily accessible on the market; PFAD, for instance, is usually classified by MSs as residue of palm oil production, and it is already utilised in biorefineries. Cottonseed oil, if sourced from contexts where it is not purified and sold for human consumption, is also classified as residue: as opposed to rapeseed and soybean, it does not spur additional demand for land.

A fifth option is represented by the use of alternative inedible feedstocks grown on marginal lands - such as *Jatropha Curcas*, *Camelina Sativa* and *Ricinus Communis* (castor). At the moment, however, EU regulations do not favour this pathway, as they do not assign them any double-counting coefficient. This fact, together with the complexity of setting up a cost-effective and large-scale supply chain, may hinder their development, although it is not to be excluded that these feedstocks may be added to part A of annex IX in reviews of the current version of RED II. The EU is also currently revising its Common Agricultural Policy, and depending on its final version, it could provide these cultivations with economic support. The EU is currently funding some minor projects with focus on these cultivations in Mediterranean regions, such as *Bio4A* and *JatroMed*; in some cases, the focus is specifically on aviation fuel [55] [56].

For the sake of completeness, algae may be mentioned as last feedstock option: in this case, however, their adoption is hindered not only by the prohibitive costs of their cultivation and harvesting, but also to their composition, which generally features a marked prevalence of unsaturated fatty acid, with consequential high hydrogen consumption [10].

Cottonseed oil can thus be added as a possible alternative in the portfolio of HVO biorefiners. Its characterisation in European legislation as residue and not as food and feed crop is beneficial for economic players, and its role may be especially important in the replacement of palm oil, given the restricted range of . Its fatty acid composition makes it suitable to hydroconversion, as well as its characterisation as residue. Real sustainability implies that such oil is not diverted from the food industry, which is the context upon which this research is built. In the next chapter, this feedstock will be analysed under an LCA perspective, to evaluate its GHG performance both under EU regulations and in comparison with other feedstock alternatives.

Cottonseed oil and chemistry of renewable diesel production

This chapter opens with a section providing a framework for cotton and its related products; it is divided in three parts, where the following topics are touched:

- Part 3.1.1 introduces the topic by outlining the history and the features of the cotton plant, its production and its processing.
- Part 3.1.2 briefly presents the many co- and by-products of the plant, highlighting their usages.
- Part 3.1.3 comments trade and usages of cotton by-products at a global level. It emerges that around 20% of global cottonseed production is not crushed, but wasted or used in the form of wholeseed. The part then concludes by overviewing the contexts that may offer some availability of seed for European and Italian biorefiners, focusing especially on Africa.

The second section in the chapter deals with the chemistry of hydroconversion, which is relevant for the development of the LCA model. The section is again constituted by three parts: these deserve a deeper glance within the topics they deal with. The first of the three, part 3.2.1, has the following functions:

- Describe chemical composition and characterisation of vegetable oils, with focus on cottonseed oil.
- Describe the processes of transesterification and hydroconversion of vegetable oils. Then, compare the two fuels - biodiesel and renewable diesel (RD): as the text points out, the latter has many advantages over biodiesel, the blending of which is also legally limited to 7% in the EU.

Part 3.2.2 is especially important for the modelling of the reactor, as it exerts the following functions:

- Describe the hydroconversion of vegetable oil; highlight the set of possible chemical reactions, their thermodynamics and kinetics.
- Discuss product yields and the effect of catalyst choice on selectivity and yield.

Part 3.2.3, which closes the chapter, deals with the specific hydroconversion process of Ecofining, exerting the following functions:

- Briefly contextualise the Ecofining process within hydroconversion processes.
- Illustrate the plant layout of a typical Ecofining plant and describe each main plant component.
- Focus on the two Italian Ecofining plants of Marghera and Gela and compare their layouts and potential. The LCA study will take into consideration the Gela refinery.

3.1 Cotton and derived products

3.1.1 Cotton plant: brief history, principles of biology and cultivation

Cotton is the most important fibre crop in the world. It belongs to the family *Malvaceae* and the genus *Gossypium*, comprehending around 50 different species. Only a handful of these has been selected for cultivation, with more than 90% of the world harvested surface covered by one single species, *Gossypium Hirsutum* - also known as upland cotton. The earliest evidence of the cultivation of plants of the genus *Gossypium* was found in Pakistan and dates back to the VI Millennium BCE. From there, the use of this plant spread in every continent, and today interests about 75 countries worldwide. [57] In Europe, its diffusion began in the early II Millennium. There, this novel industry was shaped after the model already in use in the Islamic tradition. Most of the cotton was imported from the Eastern shores of the Mediterranean, although lesser quantities were cultivated also on European soil, mainly in Greece and Southern Italy. Interestingly, Northern Italy was the set of an extraordinarily competitive industrial complex focused on the manufacturing of the raw material into cloth for export, as early as the XII Century. Milan, Cremona, Piacenza, Pavia, Bergamo were all notable examples in this value chain, with Venice and Genoa as main export hubs [58]. This early example of production complex may be considered an interesting forefather of the success of the Italian textile industry to this day.

The cotton plant is mostly cultivated as an annual crop, although it grows as a perennial plant under suitable conditions. *Gossypium Hirsutum* can reach an height of 1 to 1.5 metres, and up to 2 metres if not annually harvested. It is especially fit for tropical and subtropical climates, but has a broad diffusion in humid-warm temperate and semi-arid warm temperature zones, covering a wide range of different soils and cultural practices [26]. It thus requires high amount of solar radiation for optimal growth.

Temperature is the dominant environmental factor affecting the development of the plant: especially at an early stage, ambient temperature must average 21-22°C in order for it to develop properly [59]. This plant also necessitates regular and abundant irrigation: fields are commonly irrigated five to six times during the growing season - which lasts about 5 to 6 months [59]. In a study including all the most important cotton growing countries in the world, the usage of blue water - i.e. fresh water from surface or underground reservoirs - was quantified to an average of 1300 m^3 per ton of fibre. This value registered large variations from country to country, witnessing the wide varieties of climates and practices in cotton cultivation [60]. Another important factor influencing cotton yield and quality is its sensitivity to abiotic stress, i.e. to droughts, waterlogging and sudden temperature variations.

Once grown, the plant shows a marked vertical development, with flat, pointy leaves that can orient to maximise sunlight absorption. Some of the branches carry flowers, called buds, which in a matter of 3 days after blooming fall, uncovering green pods called cotton bolls. These bolls contain both seeds and cotton lint. They are initially wrapped in leaves, which then gradually open at maturity of the plant.

The harvesting of these bolls can be carried out either manually or with the help of cotton pickers. In the latter case, it is common to employ a class of chemicals called defoliants, which open the boll and thus facilitate the separation of seed cotton from the stem. The use of agrochemicals can be particularly intense in the cultivation of cotton: fertilisers, insecticides, herbicides, growth regulators and defoliants are frequently employed, consuming 11% of the world's agrochemicals [61]. Once harvested, the seed cotton must be subject to the separation of the lint from seeds and solid residues. The invention of the cotton gin represented a breakthrough discovery for this industry, and its introduction marked the onset of a widespread adoption of cotton as textile. Eli Whitney invented the first example of cotton gin in 1793: his machine could process in only 30 minutes the amount of fibre separated by the hands of an operator in one entire day [57]. In a ginning factory, seed cotton first undergoes the removal of trash, which represents some percentage points by weight and is especially present in machine-harvested cotton. The remaining material is composed by lint and seeds, which are present in the proportion of 35% and 65% respectively [62]. After ginning, cotton lint is ready to be spun and turned into cotton cloth.

3.1.2 Products and by-products of the cotton industry

Cotton features a variety of products and by-products, with a great potential for circular economy practices and value addition. A cotton gin generates three main outputs, namely cotton lint, cottonseed and cotton trash. Cotton lint and cottonseeds are the two main co-products of the ginning process, with lint being the most important one in economic terms, thanks to its high value in many industries - primarily in the textile sector. Cotton trash is made up of leaves and stalk fragments, dust and other residues, which together constitute about 10% of fresh seed cotton weight when machine-harvested. This material can be used as feed, burned for energy or possibly even pressed into fuel pellets [63].

Cotton fibre is by far the most utilised natural fibre by production volume, representing more than 75% of the market share. When including artificial fibres into the analysis, production of cotton covers 24.4% of the global demand [64]. The use of cotton lint ranges from the production of clothes and house items to disposable items for personal and medical care.

Cottonseed is the most abundant product by weight. Its oil content typically ranges between 16 and 20%, with variations depending mainly on the species [65], but higher oil contents are also reported. It is an ovoid weighing about 1 dg, covered in short cellulosic hairs called linters. In the form of wholeseed, it finds application as animal feed, and specifically as feed for adult cattle. The main issue in such use is its gossypol content, as this phenolic compound is toxic for most beings: its ingestion can lead to acute poisoning, although its relatively low concentration in nature most frequently leads the animal to a gradual health deterioration. The consequences include respiratory distress, anorexia, weakness, and death. Since monogastric animals are more exposed to gossypol poisoning than ruminants, cottonseed is mostly fed to adult cows only [66]. Cottonseed is considered a high-quality feed by cattle farmers, since it combines a high caloric content with a remarkable protein content.

In many contexts, cottonseed is delivered to an extraction plant, in order to fraction the seed into its constituents: oil, meal, hulls and linters. This process can be carried out either by simple physical pressing, or by also exploiting solvent extraction. The typical composition of raw cottonseed by weight is: 45% meal, 27% hulls and 8% linters, with the rest being oil and moisture [62]. When solvent extraction is employed, the yield of oil is maximised to 95% and more, minimising the presence of residual oil in the cottonseed meal. All the products of the extraction facility represent a potential source of revenues.

The oil, after refining and solvent extraction specifically targeted at removing gossypol, is valuable as cooking oil. Its sale is frequent in many cotton-producing countries, due to its relatively low price and good flavour stability: in China, for instance, cottonseed oil represents 8.9% of domestic edible oil production [67]. As for many other vegetable oils, cottonseed oil finds application also in other industries, such as in the production of soap.

Cottonseed meal is normally utilised as cattle feed. Similar in all respects to whole cottonseed, it is regarded as a valid protein feed supplement: in terms of protein content, 1 kg of average cottonseed meal is equivalent to 0.81% kg of soybean meal; all other common meals, by contrast, exhibit lower protein content, as 1 kg of rapeseed meal and sunflower seed meal would equal 0.71 kg and 0.66 kg of soybean meal respectively [20]. With respect to wholeseed, the only downside of the meal is its lower energy content, because of its low to negligible oil content.

Cottonseed hulls are typically removed from the wholeseed via a dehulling machine and then mixed with the meal, in order to enrich its fibre content. In alternative, they can be burnt *in loco*, together with the trash, to supply carbon-neutral heat to the oil extraction process. Cotton linters may be left on the hulls or removed prior to seed dehulling and crushing. In the former case, lintered hulls can be mixed with the meal and sold as feed; in the latter case, prior additional investment in the extraction plant, the linters can be profitably sold to many niche market segments.

Finally, it is relevant to mention cotton stalks as a potential additional source of value added. It is often left on field after harvest and then ploughed underground, with a fertilising effect for the next crop. Some countries have adopted more innovative approaches, using it to produce fuel pellets or as a substrate for the growth of edible mushrooms [67] [68].

It is important to highlight the two characteristic features of cottonseed when compared to all other common sources of vegetable oil in the market, such as palm, soybean and rapeseed oil. Differently from these sources, in fact, cottonseed is not the main product of a value chain, but represents instead a residue of an industry whose main goal is the production of cotton lint. The second important difference is the toxicity of seed and oil in cotton, which hinders its use as it is and requires additional processing in most cases.

3.1.3 Global market and trade of cottonseed and related subproducts

The bulk of global cotton production is concentrated in a restricted number of countries. as shown in figure 3.1: more than half of the world cotton production, which amounted to 121.7 million bales (about 26500 kton) depends on only three countries, namely India, China and the USA [28]. Notwithstanding, this industry interests a remarkably large number of countries with a tiny market share of less than 1% of global volumes; furthermore, many belong to the category of developing countries: about 75 out of a total of 90 countries, in fact, belong to this category [26].

In general, cotton cultivation features the prevalence of small, family-run businesses, where hand-picked cotton is preferred over machine-harvested cotton: at present, only 30% of the world production relies on machine harvesting [57]. This feature is especially marked in developing nations: here, labour-intensive activities prevail, with cotton representing an important driver of economic growth and wealth distribution in many rural areas. In countries like the United States, Australia and Brazil, cotton growers retain ownership of the crop after ginning; in many countries, seed cotton is typically bought by traders or ginning companies at the farm level, thus transferring the ownership of the good [62]. There are consistent cross-country differences in product yield, mirroring the different growth conditions and agricultural practices: Australia holds the record for the highest seed cotton yields, with 5416 kg/ha, while the lowest average yield has been recorded in Africa and amounts to just 912 kg/ha [57].

The production of cottonseed and its related subproducts - i.e. oil, meal, hulls and linters - is clearly dependent on the status of the cotton fibre market. Production has

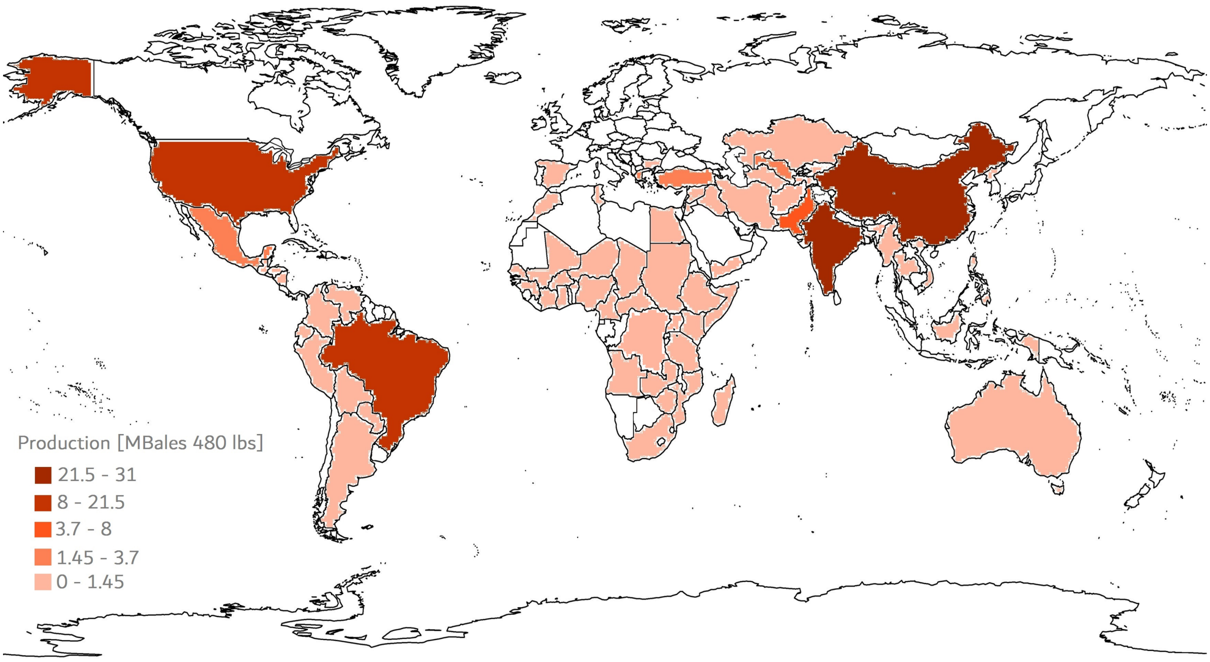


Figure 3.1: Cotton production by country in harvest year 2019/2020 [28].

recently recovered from an acute production drop in 2015; the causes of such contraction have been envisaged in adverse weather, high stocks and lower global demand due to competition with synthetic fibres, whose price has been decreasing together with the decrease of crude oil price. In the next years, a mild production growth of 2% p.a. is expected, and, by 2025, India may become the first producer by volume [69].

The United States Department of Agriculture (USDA) regularly collects data on the handling of cottonseed at a global level. Harvest year 2018/2019 yielded 43.4 Mt of seeds, of which 33.52 Mt underwent crushing. Admitting a small portion of seeds - approximately 2 to 5% [57] - has to be kept for sowing the next harvest, the remaining uncrushed seed constitutes about 20% of global production [20]. In the most favourable contexts, e.g. in Israel, such cotton wholeseed may be sold as cattle feed, but it may also well become waste: this is especially the case for countries that do not feature a modern cotton industry with a solid supply chain, as for many cotton-producing African countries [21]; upgrading the use of cottonseed, independently from the specific application, can thus be a powerful driver of development. If such upgrade went completely towards biofuel production, such uncrushed seed would globally yield about 1300 kton of oil, which would cover about 50% of EU HVO production as of 2017 [6]; this corresponds to covering about 42% of global HVO production [22].

Since this research focuses on RD production in EU and, more specifically, Italy, it is appropriate to now restrict the scope to the cotton producers in the Mediterranean. As shown in figure 3.1, cotton is produced in numerous Mediterranean countries, Greece and Spain being the only EU countries to cultivate it to some extent; an appreciable production is also present in the Middle East. To give an idea of the production volume, devolving the entirety of cottonseed production from Greece and Egypt to oil for RD would cover about 80 to 90% of Italian feedstock demand [20]. However, gathering country-specific data about current cottonseed uses is hard for most countries, implying that it's not possible to accurately estimate the portion of uncrushed cottonseed in their production. In the Mediterranean, Turkey is the most important cotton producing country, placing seventh in the world for cotton production by volume [28]. In this country, however, a robust cottonseed-oil market exists: it is thus unlikely that using Turkish cottonseed for biofuels production would improve its sustainability, rather decreasing it.

Subsaharan Africa is a very interesting context for a possible use of cottonseed oil for biofuels production. Nearly all of the countries in the region produce cotton, and there is sufficient evidence for a large portion of cottonseed being used uncrushed or even wasted. As shown in figure 3.2, many countries do not properly harness their potential in cottonseed [21].

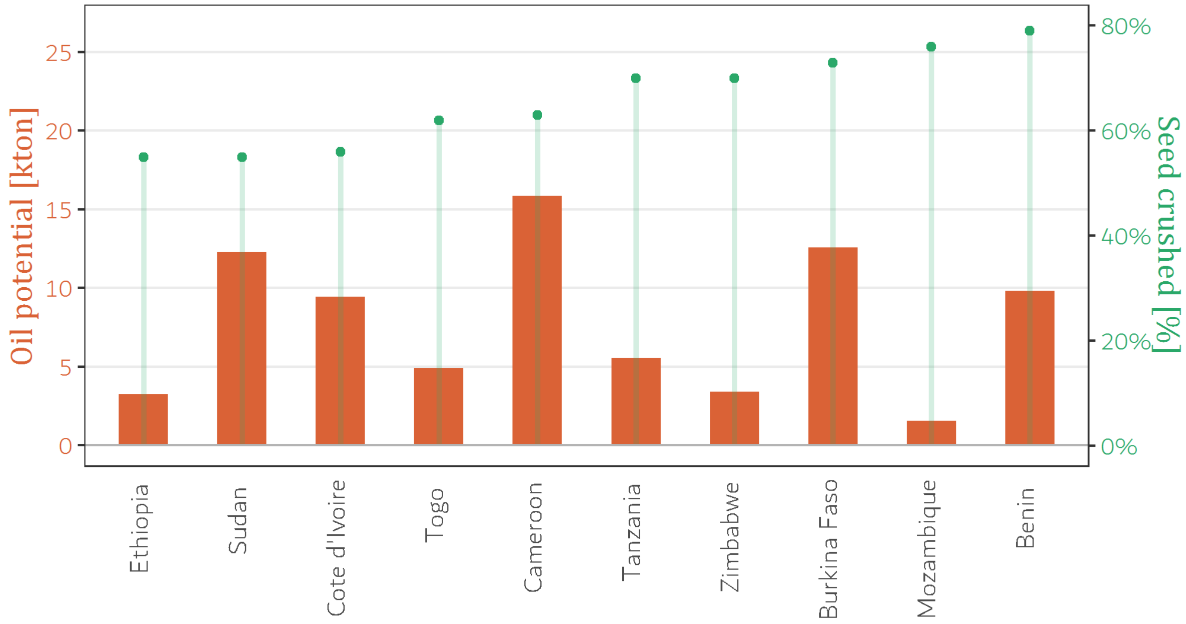


Figure 3.2: Portion of crushed cottonseed in selected African countries, and oil potential of this uncrushed cottonseed [21].

The figure shows the top ten countries with the least percentage of crushed seeds in their economy. These countries have a portion of crushed seeds of 80% or less, with three of them crushing less than 60% of their seed. If crushed, this amount of seed could cover around 20% of 2017 Italian RD feedstock demand [6]. Five of these countries alone - Benin, Cameroon, Côte d'Ivoire, Burkina Faso and Sudan - represent more than 75% of this amount.

In order to upgrade their cottonseed subproducts industry, some African countries have instituted governmental bodies for the development and the upgrade of their cotton industry, even by means of sectorial cooperation between States - as for Benin, Burkina Faso, Mali and Chad [68] [70] [71]. Cottonseed has also recently gained the attention of international projects focusing on the African continent: the United Nations Conference on Trade and Development (UNCTAD) and the World Trade Organisation (WTO) are involved in a number o projects interesting Tanzania, Uganda, Zambia and Zimbabwe [21].

This UNCTAD project highlights a wide range of uses for cottonseed subproducts that could benefit local industries, as reported in figure 3.3.

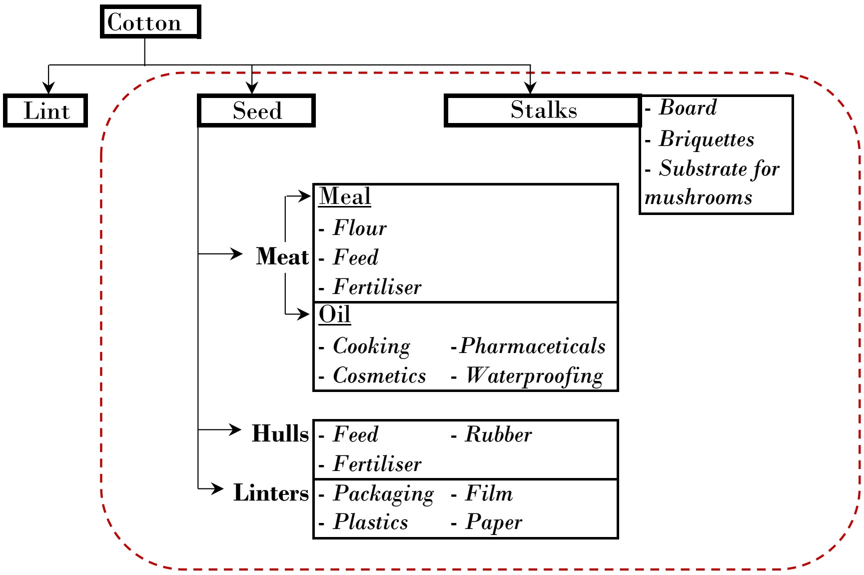


Figure 3.3: Focus area of UNCTAD project: "Promoting cotton by-products in Eastern and Southern Africa" [29].

The focus is on the food and feed industry, with innovative uses also for cotton stalks such as mushroom cultivation, which is already practised in China [67]; another beneficial possibility for stalks is briquettes production. These solutions go into the direction of solving the issues of food insecurity and indoor pollution, while providing farmers with increased sources of revenues.

3.2 The hydrogenation of vegetable oils

3.2.1 Overview and comparison of renewable diesel with biodiesel

Oils and fats represent a major feedstock for the production of biofuels. Their processing mainly delivers fuels in the range of diesel and kerosene, which are typically blended in commercial mixtures to lower the usage of fossil fuels. At a global scale, the production of vegetable oils account for a share of about 80%, and fats of animal origin for the remaining 20%. Most of this production is destined to be used as food or animal feed, whereas the production of biofuels covers an estimated 14% share of the total amount [10].

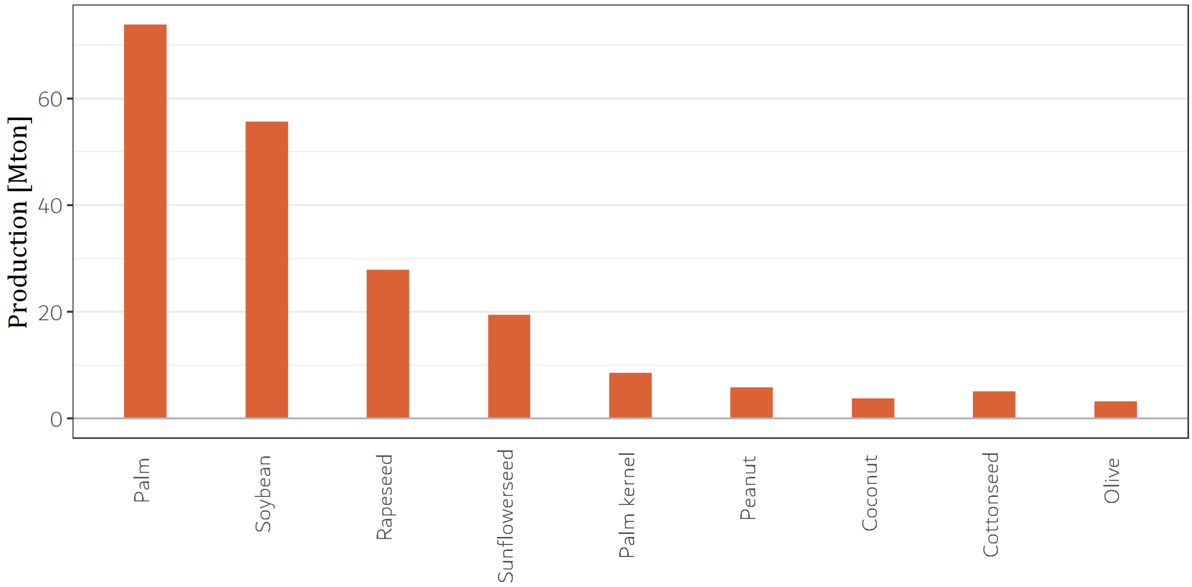
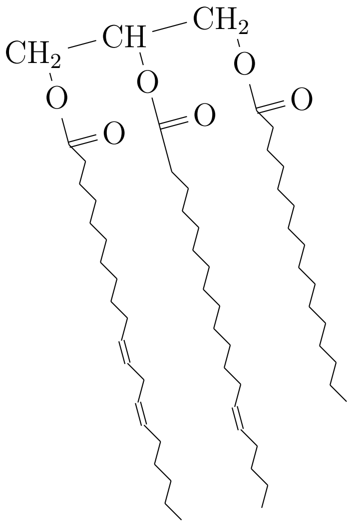


Figure 3.4: Major vegetable oils by global production in market year 2018/2019, in commodity view [20].

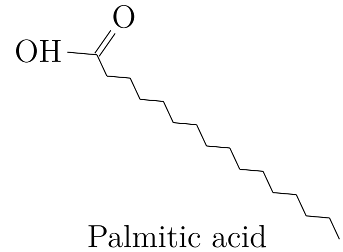
Figure 3.4 shows the most relevant vegetable oils in the global mix: it can be seen that palm oil - including palm kernel oil - and soybean oil account together for an impressive 68% of the total. Cottonseed oil ranks seventh, with a yearly production of about 5 Mt [20].

All vegetable oils consist primarily of a class of esters called triglycerides. The building blocks of a triglyceride molecule are three fatty acids and one molecule of glycerol. Figure 3.5 shows an example of triglyceride and its constituent fatty acids. Two properties characterise a fatty acid: the length of its carbon chain and the number of double bonds (olefinic bonds), i.e. the degree of unsaturation. The molecule of palmitic acid shown in the picture, for instance, features a chain of 16 carbon atoms and no unsaturations.

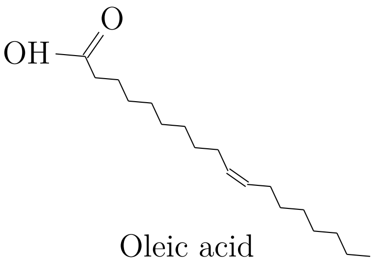
In vegetable oils, concentration of triglycerides typically ranges between 95% and 97% by weight; the remaining 3-5% is constituted other compounds, such as Free Fatty



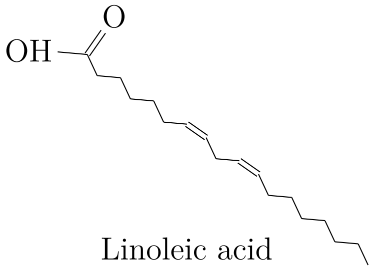
A triglyceride



Palmitic acid



Oleic acid



Linoleic acid

Figure 3.5: Representation of a triglyceride molecule and its constituent fatty acids.

Acid (FFA), phospholipids and other organic impurities. In addition, they contain minor concentrations of inorganic materials, such as chlorine, alkali, alkali earth metals and transition metals.

Vegetable oils are classified based on the two aforementioned properties, i.e. carbon-chain length and degree of saturation. In table 3.1, the most common vegetable oils are broken down to their typical fatty-acid composition.

From the table, it can be noticed that all fatty acids feature an even number of carbon atoms. In addition, most vegetable oils feature a marked prevalence of fatty acids in the range of 14-16 and 18-20 carbon atoms - with the exceptions of peanut and palm kernel oil. This carbon-chain length range matches the one of common diesel fuels. It is possible also to compare vegetable oils based on their degree of saturation: for instance, palm oil and coconut oil feature very high degrees of saturation. For this reason, differently from the other oils in the table, they are semi-solid at room temperature, much like animal fats.

Vegetable oils such as cottonseed oil could, in principle, be directly employed as transportation fuel - in what is known as Straight Vegetable Oil (SVO). Interestingly, the original diesel engine was capable of running both with mineral oil and with vegetable oil. In front of the international audience at the Paris International Exposition of 1900, Rudolf

Table 3.1: Fatty acid composition of selected vegetable oils,in percentage weight. [10]

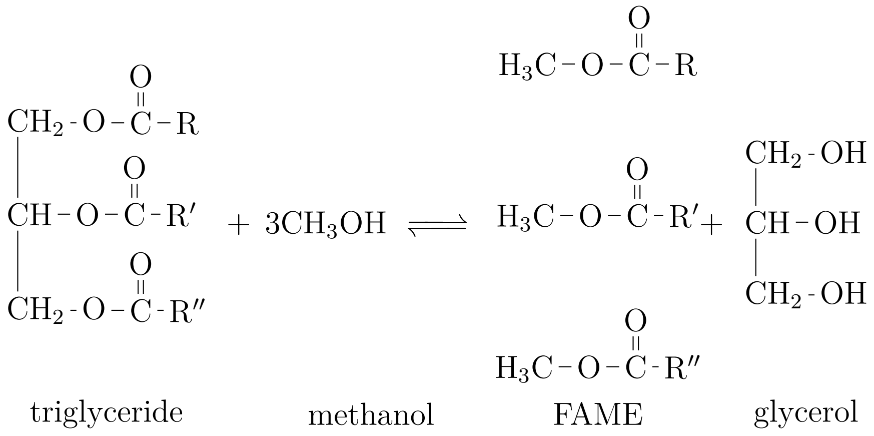
	Fatty acids						
	Mystiric $C_{14:0}$	Palmitic $C_{16:0}$	Stearic $C_{18:0}$	Oleic $C_{18:1}$	Linoleic $C_{18:2}$	Linolenic $C_{18:3}$	Other
Palm	1	44	4	40	10	Traces	1
Soybean	-	10	4	23	53	8	1.5 $C_{20\text{-}22}$
Rapeseed	-	5	2.5	59	21	9	2 $C_{20\text{-}22}$
Sunflower	-	6	5	18	69	<0.5	1.5 $C_{20\text{-}22}$
Palm kernel	16	8	3	16	2	-	54 $C_{8\text{-}12:0}$
Cottonseed	1	25	2	18	53	0.3	0.7 $C_{16:1}$
Coconut	18	9	3	6	2	Traces	62 $C_{8\text{-}12:0}$

In $C_{x:y}$, **x** stands for the C-chain length, **y** for the number of double bonds.

Diesel used peanut oil as fuel for his invention [72]. The core issue linked to the use of SVO is its viscosity, one order of magnitude higher than that of mineral-sourced diesel fuel. This high viscosity causes poor atomization of the fuel in the combustion chamber, ultimately resulting in solid carbon deposits and clogging. Its physical characteristics make SVO sensitive to low temperatures, with a cloud temperature that is too high to make its use feasible in many countries, especially in the upper Northern Hemisphere.

In order to reduce the viscosity of SVO and improve its combustion properties, two alternative processes are commonly employed: transesterification and hydroconversion. These are the two pathways leading from oils and fats to biofuels as of today, which will be briefly analysed and compared in the next paragraphs.

Transesterification is defined as the chemical reaction of an ester with an alcohol, delivering a different alcohol and a different ester. This reaction finds application in many fields, but in the context of biofuel from vegetable oils, it typically involves triglyceride and methanol as reactants, delivering glycerol and fatty acid methyl ester (FAME) as products. This general reaction is characterised by the stoichiometry reported in 3.1: one mole of triglyceride and three moles of methanol produce three moles of FAME and one mole of glycerol.



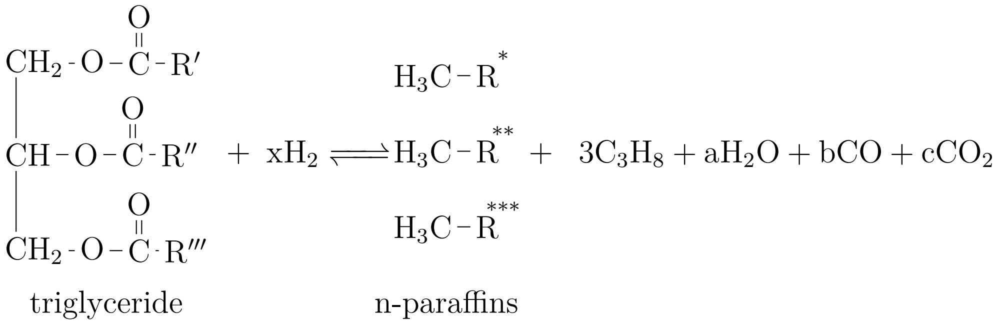
$$\begin{array}{l} CH_2\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R \\ | \\ CH\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R' \\ | \\ CH_2\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R'' \end{array} + 3CH_3OH \rightleftharpoons \begin{array}{l} H_3C\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R \\ \\ H_3C\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R' \\ \\ H_3C\text{-}O\text{-}\overset{O}{\overset{\|}{C}}\text{-}R'' \end{array} + \begin{array}{l} CH_2\text{-}OH \\ | \\ CH\text{-}OH \\ | \\ CH_2\text{-}OH \end{array} \tag{3.1}$$

The reaction is substantially an equilibrium process: it ensures high yield and feasibility even at ambient conditions [73]. In commercial reactors, it is commonly carried out in the presence of an alkaline catalyst in homogeneous phase, under ambient pressure and temperatures in the range of 40 to 70°C [74] [75] [76]. This process can be easily operated at small scale, typically employing batch reactors. Furthermore, it does not alter the nature of fatty acids in the triglyceride: its only aim is the reduction of viscosity, while the degree of saturation of fatty acids remains unaltered. FAME is the finished product - normally referred to as biodiesel - and it can be blended in commercial fuels; glycerol is a by-product, and a demand thereof exists in some markets. Notwithstanding, a large excess of supply is currently characterising its marketability.

The term hydroconversion defines a process involving the consumption of hydrogen to treat a feedstock. In the form of hydrotreatment, it has been in use for decades in the refining of mineral oil, where its main purpose is the removal of heteroatoms - primarily sulphur, in Hydrodesulphurisation (HDS) units. Some companies have also adapted their HDS units to be co-fed with a stream of vegetable oil, but this co-feeding presents a number of limitations: sulphur removal becomes less effective in the presence of oxygen, requiring harsher reaction conditions; catalyst life is compromised by the presence of oxygen; the treatment of additional by-products [10]. The alternative is a stand-alone unit, which, although more capital-intensive, eliminates the aforementioned issues.

In the case of vegetable oils, the essential purpose of hydroconversion is the cleavage of oxygen bridges in the triglyceride, together with the saturation of olefinic bonds along their carbon chains, in order to deliver n-paraffins (linear paraffins); the reactants are thus simply the triglycerides in vegetable oil and hydrogen.

Equation 3.2 portrays the general reaction of vegetable oil hydrotreatment. Three different chemical reactions can occur at this stage: depending on the prevalent reaction, there will be differences in the side products; however, as outlined in the equations, any combination of these reactions will always deliver three moles of linear paraffins and one mole of propane for each mole of triglyceride, according to stoichiometry. The characteristic of each reaction pathway will be analysed in detail in section 3.2.2.



$$\begin{array}{l} \mathrm{CH_2{-}O{-}\overset{\overset{\displaystyle O}{\|}}{C}{-}R'} \\ | \\ \mathrm{CH{-}O{-}\overset{\overset{\displaystyle O}{\|}}{C}{-}R''} \\ | \\ \mathrm{CH_2{-}O{-}\overset{\overset{\displaystyle O}{\|}}{C}{-}R'''} \\ \text{triglyceride} \end{array} + xH_2 \rightleftharpoons \begin{array}{l} \mathrm{H_3C{-}R^{*}} \\ \mathrm{H_3C{-}R^{**}} \\ \mathrm{H_3C{-}R^{***}} \\ \text{n-paraffins} \end{array} + 3C_3H_8 + aH_2O + bCO + cCO_2 \qquad (3.2)$$

Subsequent to hydrotreatment, hydroconversion of vegetable oils typically features a step of hydroisomerisation of the n-paraffin, with the sole purpose of improving cold flow properties, i.e. decreasing freezing point and cloud point: the end product is thus an iso-paraffinic mix in the range of diesel fuel, with minor yield of lighter hydrocarbons and propane. This process is carried out in presence of a solid catalyst and at temperatures and pressure substantially higher than in the transesterification process, in the range of 270 to 350°C and 2 to 7 MPa respectively [10].

The presented general characterisations of transesterification and hydroconversion allow now for a comparison between the two processes in terms of versatility, product properties and applications. Transesterification has a first important shortcoming in its lower feedstock flexibility: due to the fact that olefinic bonds in the fatty acid remain unaltered, physical properties of the product FAME resemble the properties of the starting oil. In case of highly saturated vegetable oils, this implies poor cold properties of the resulting fuel, as double bonds help contain the freezing temperature. On the other hand, an excess of olefinic bonds in the feedstock leads to a FAME with low oxidative stability, which hinders the possibility to store the fuel before use. The properties of a HVO are instead largely feedstock-independent, thus allowing for more flexibility. Hydroconversion can also process oils rich in FFA without complications, whereas alkaline-catalysed transesterification favours soap formation by combination of the catalyst with these FFAs, ultimately lowering the yield and complicating product separation [74]. Conversely, hydroconversion is sensitive to the presence of olefinic bonds in terms of hydrogen consumption, which represents - together with feedstock cost - the most relevant operating cost in the unit. The second class of shortcomings of biodiesel consists of its inferior final product properties if compared to HVO. Besides the already mentioned poor oxidative stability, biodiesel feature a lower heating value and a lower cetane number than HVO, as shown in table 3.2. In the case of HVO, these two parameters are instead higher than in petroleum diesel, thus making it a premium fuel.

Given all the limitations of biodiesel, European regulations set a blending threshold of 7% in conventional diesel fuels. The compliance with biofuel targets thus requires a further expansion of HVO production, on which this research focuses. This and other implications of new European regulations are discussed in section 2.

Table 3.2: Selected physical properties of ULSD, FAME (biodiesel) and a typical HVO diesel fuel. [11]

	ULSD	**FAME**	**HVO diesel**
Oxygen %	0	11	0
Specific gravity	0.84	0.88	0.78
Sulphur [ppm]	<10	<1	<1
LHV [MJ/kg]	43	38	44
Cloud point [°C]	-5	-5 to +10	-20 to+20
CN	40	50 to 65	70 to 90
Stability	Good	Marginal	Good

3.2.2 Chemistry of the production of renewable diesel

In order to yield high-quality transportation fuel, the hydroconversion of triglycerides requires two stages in series: each of these is characterised by a specific function, specific reaction parameters and specific catalyst.

In the first stage, double bonds are saturated and the triglyceride molecule is cracked; this step yields a number of products, the most relevant of which is constituted by linear paraffins. In order to improve the cold flow properties, a second stage for the isomerisation of these n-paraffins is needed. Both stages are carried out under hydrogen pressure: in the first phase, hydrogen is needed both in triglyceride cracking and in bond saturation, whereas the second stage consumes hydrogen only in the case of paraffin cracking, and no consumption is associated to isomerisation, which is a mere reorganisation of the molecular structure. A simplified representation of the first deoxygenation stage, highlighting feed and products, is reported in figure 3.6.

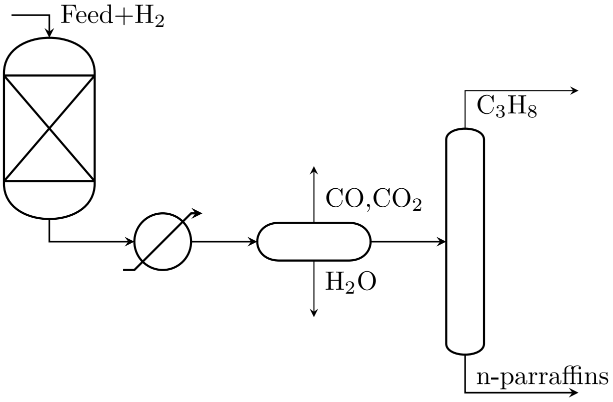


Figure 3.6: Schematic representation of the deoxygenation stage and its products (reactor section - condenser - settler - fractionation section).

At this stage, vegetable oil - which is normally consumed at 3 to 3.5 % weight fraction, depending on the feed - is mixed with hydrogen, ultimately delivering a set of different products: primarily n-paraffins, but also propane and other lighter hydrocarbons in lesser quantities, H_2O, CO and CO_2. The overall reaction is highly exothermic and characterised by highly favourable kinetics, as experiments have led to complete conversion for short contact times and moderate temperatures, i.e. above 310°C [33].

The relative abundance of H_2O with respect to CO and CO_2 is determined by the prevalence of one of the three possible reaction pathways for the removal of oxygen in the cracking of triglycerides: hydrodeoxygenation (HDO), decarbonylation (DCO) and decarboxylation (DCO2). These reactions are reported in equations 3.3a, 3.3b, 3.3c; in these examples, the sample feed is a triglyceride with only C_{18} fatty acids, as for instance oleic or linoleic acid.

$$\mathrm{C_{57}H_{(110\text{-}2x)}O_6 + (12 + x)H_2 \rightleftharpoons 3\,C_{18}H_{38} + C_3H_8 + 6\,H_2O} \tag{3.3a}$$

$$\mathrm{C_{57}H_{(110\text{-}2x)}O_6 + (9 + x)H_2 \rightleftharpoons 3\,C_{17}H_{36} + C_3H_8 + 3\,CO + 3\,H_2O} \tag{3.3b}$$

$$\mathrm{C_{57}H_{(110\text{-}2x)}O_6 + (3 + x)H_2 \rightleftharpoons 3\,C_{17}H_{36} + C_3H_8 + 3\,CO_2} \tag{3.3c}$$

As shown in the reactions, stoichiometric hydrogen depends on a quantity x, which stands for the total number of olefinic bonds in the triglyceride molecule. In all three reactions, the main co-products are a paraffinic mixture and propane, in an ideal molar proportion of 3 to 1 - in case of a feed consisting purely of triglycerides.

The distribution of n-paraffins with regards to carbon-chain length mirrors the composition of the vegetable oil feed, with an important difference between HDO and DCO/DCO2: while the first reaction does not alter the carbon-chain length of the triglyceride, the latter two reactions deliver paraffins with an odd number of carbon atoms, as one carbon leaves in the form of carbon oxide. Consequently, the presence of DCO/DCO2 lowers the yield by weight. This phenomenon is accompanied by a lower consumption of hydrogen in the cracking of the triglyceride; as the reactions show, a maximum of 12 moles per mole of triglyceride in the case of HDO, and a minimum of 3 moles in the case of DCO2.

Based on the aforementioned considerations, the prevalence of a certain pathway is key for determining two crucial reaction variables: paraffinic yield and hydrogen consumption. There are two main levers influencing which reaction is favoured: the tuning of thermodynamic parameters and the choice of catalyst.

In fact, all three reactions are exothermic, but their standard reaction enthalpies - which change depending on the specific triglyceride - are strongly different: DCO2 is less exothermic than HDO by a factor of 7. Thus, higher temperatures lead to higher selectivity towards DCO and DCO2. Also pressure exerts an effect: higher pressure favours HDO over the other pathways [33].

The factors influencing the reaction pathways are not only of thermodynamic nature, but also linked to kinetics: in this respect, the role of catalyst composition is fundamental. Generally, the main catalyst used for HDO corresponds to the ones in use in other hydrotreating processes, i.e. a sulphide of a transition metal - typically molybdenum or

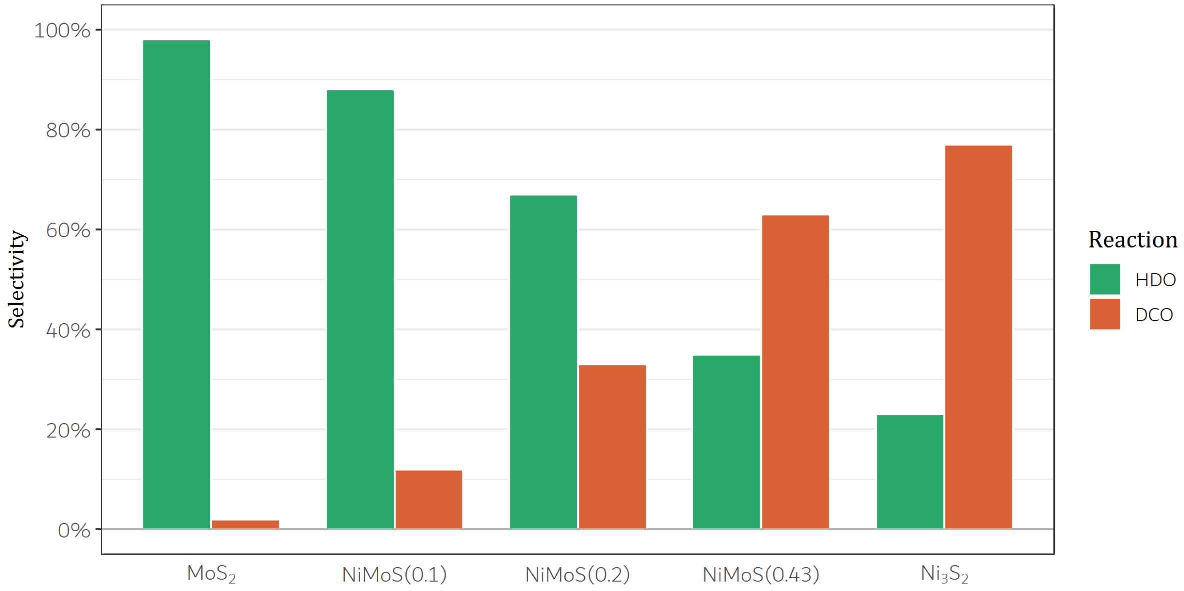


Figure 3.7: Hydrogenation reaction selectivity depending on catalyst combination [10].

tungsten. Common support materials include alumina, silica-alumina and zeolites - here in order of increasing acidic property. This catalyst is usually accompanied by a second transition metal - typically cobalt or nickel - acting as a promoter [10]. Experimental trial has proved that the relative prevalence of molybdenum sulphides or nickel sulphides, representing the two classes of catalyst employed, determines the selectivity towards a certain reaction. The experiments involved a heptane feed and a variety of unsupported sulphided catalysts: MoS_2, Ni_2S_2, and Ni-promoted MoS_2 in different Ni/Mo ratios.

The results are shown in table 3.7: HDO is favoured by group VI metals like molybdenum, whereas nickel favours DCO/DCO2. Molybdenite (MoS_2) is thus characterised by the highest selectivity towards HDO, while Ni_2S_2 by the highest selectivity towards DCO/DCO2 [77]. As pointed out in the previous section, the Ecofining process adopts a Ni-Mo catalyst, although the exact proportions are undisclosed to the public. Nonetheless, it is reasonable to assume a selectivity for HDO S_{HDO} in the range of 35 to 85%.

Regardless of the main reaction pathway, the resulting n-paraffinic product has the problem of gelification at low temperatures, i.e. high cloud temperature: for this reason, n-paraffins are subject to a second stage, with the purpose of improving its cold-flow properties by branching the paraffinic molecules. In this stage, hydrogen consumption is very low, and it is solely associated with hydrocracking reactions, that to a certain extent are inevitable. An example of the two classes of reactions occurring at this stage, isomerisation and hydrocracking, is shown in figure 3.8.

The absence of heteroatoms in the feed allows to use noble metals as catalysts: platinum and palladium, in fact, are sensitive to heteroatoms poisoning. This enhances the activity compared to transition metals and facilitates the process.

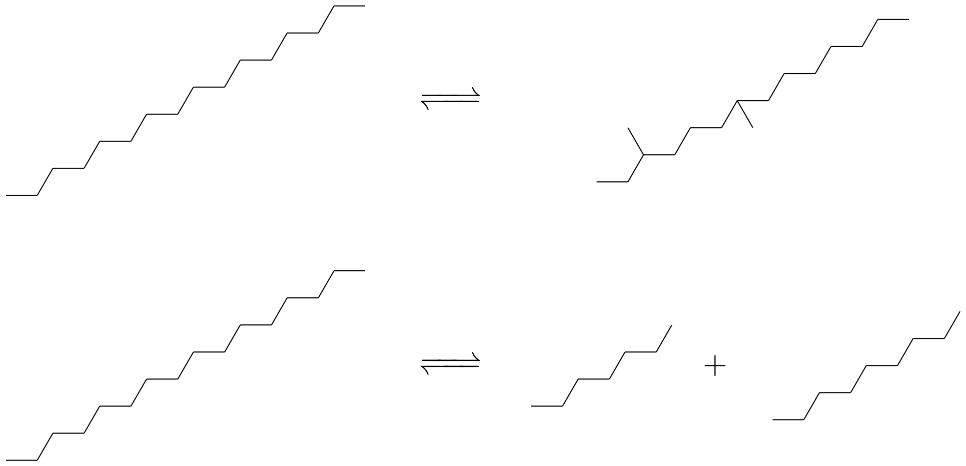


Figure 3.8: Examples of isomerisation and cracking of octadecane.

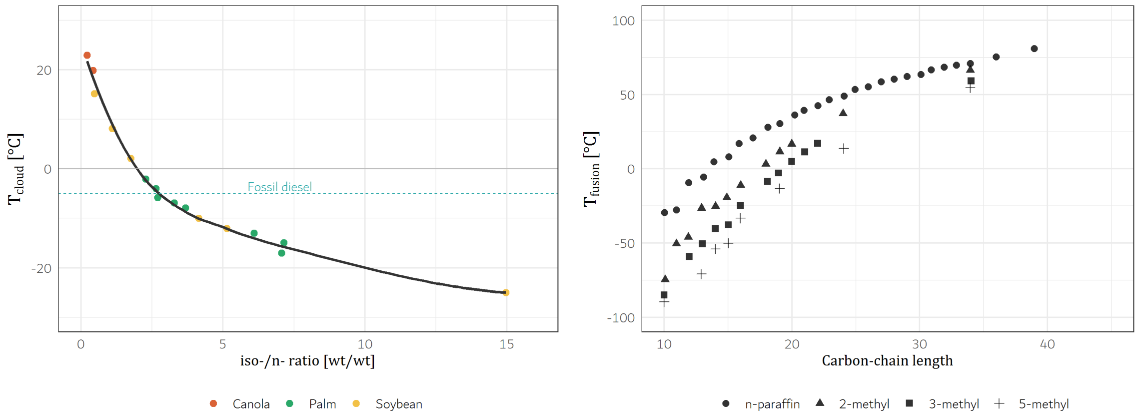


Figure 3.9: Effect of branching and carbon-chain length on cold-flow properties. Adapted from [10].

Branching has a vigorous effect on cold-flow properties: figure 3.9 shows how the reduction in cloud point is proportional to the branching degree and to the branching complexity, and inversely proportional to carbon-chain length [10]. Increasing the process severity by increasing temperature can lead to cloud points as low as -20°C. However, an increase in temperature is accompanied by hydrocracking reactions, which lower the diesel yield while increasing naphtha and light hydrocarbons production; this phenomenon is undesired unless the process aims at producing renewable jet fuel.

At fixed final cloud temperature, palm oil requires less isomerisation than most other vegetable oils, thanks to the shorter average length of its fatty acids; in any case, some degree of isomerisation is always desired prior to mixing with fossil diesel, whose cloud temperature is typically around -5°C [10].

Isomerisation is not only associated with better cold-flow properties, but also with undesired reductions in cetane number and heating value; however, these side-effects do not compromise the quality of the end product, which is still a premium fuel if compared to conventional diesel. As already pointed out, this stage can be set to deliver jet fuel as main product: in this case, more severe temperature conditions are adopted. Acidity of the catalyst support is also of importance to enhance cracking reactions: for this reason, zeolites are preferred over amorphous silica-alumina. The final yields of this process in diesel oil mode are reported in table 3.3 [10].

Table 3.3: Typical yields of Ecofining in diesel oil mode [33].

	Stream	**Feed and Yields** [$wt\%$]
Feed	Vegetable oil	96.5-97
	Hydrogen	3-3.5
Products	Diesel	75-85
	Naphtha	1-8
	C_3H_8	4-5
	H_2O	6-8
	$CO+CO_2$	3-4

Diesel yield is high, between 75 and 85%; naphtha yield has a wide range of variation, and it will normally be higher when better cold-flow properties, i.e. lower cloud point, are desired; propane and light hydrocarbons are mainly linked to the first stage, and can vary from 4 to 5% by weight; the quantities of the other by-products depend on the conditions in the first stage [10].

3.2.3 The Ecofining process

Since hydroconversion of vegetable oil is a fairly novel development, HVO technologies are not numerous, and they are normally covered by patents. The Finnish company Neste Oil is one of the pioneers in the field, with its proprietary process, NExBTL: it opened its first facility in 2007, and has since then become the largest HVO producer in the world, with a current 2.7 Mt of renewable fuel production and planned expansions bringing total capacity to 4.5 Mt by 2022. [78] In 2004, the Danish Haldor Topsøe has developed its own technology, known as HydroFlex, where the emphasis is put on feedstock flexibility [79]. The list of existing technologies continues with Vegan, developed by the French company Axens Group; Bio-Synfining, created by the American Syntroleum Corporation; UPM BioVerno, tailored to convert tall oil into RD [80]. Eni and Honeywell UOP developed a process known as Ecofining. The Italian energy company Eni and the American Honeywell UOP started a collaboration in 2005 for the development of a hydroconversion technology that took the name Ecofining. Since its introduction, it has found application in Italian and American biorefineries alike, with raising interest from other energy companies, such as the Chinese major CNPC [33] [81].

In Italy, the first commercial plant was obtained by converting two existing hydrodesulphurisation units in the Venice Marghera conventional refinery. This was seen as a way to cut on capital expenditures compared to *ex novo* construction while revamping an asset that was considered outdated before the intervention, as the refinery start-up dates back to 1926. Its operation started in 2014, with a processing capacity of 360 kt/y of vegetable oil; the company has programmed an upgrade which should take its capacity to 600 kt/y by 2021. More recently, Eni has invested in a 750 kt/y new stand-alone unit in Gela, Sicily, thought to replace the existing petrochemical plant. The production began in 2019. [82] These plants are thought to be complemented with a pretreatment unit, in order to improve feedstock flexibility and allow to expand the feedstock pool beyond refined virgin vegetable oils.

Ecofining is designed as a two-stage stand-alone process. The core of the process is a series of two reactors, whose primary tasks are deoxygenation and isomerisation respectively. The process scheme is shown in figure 3.10.

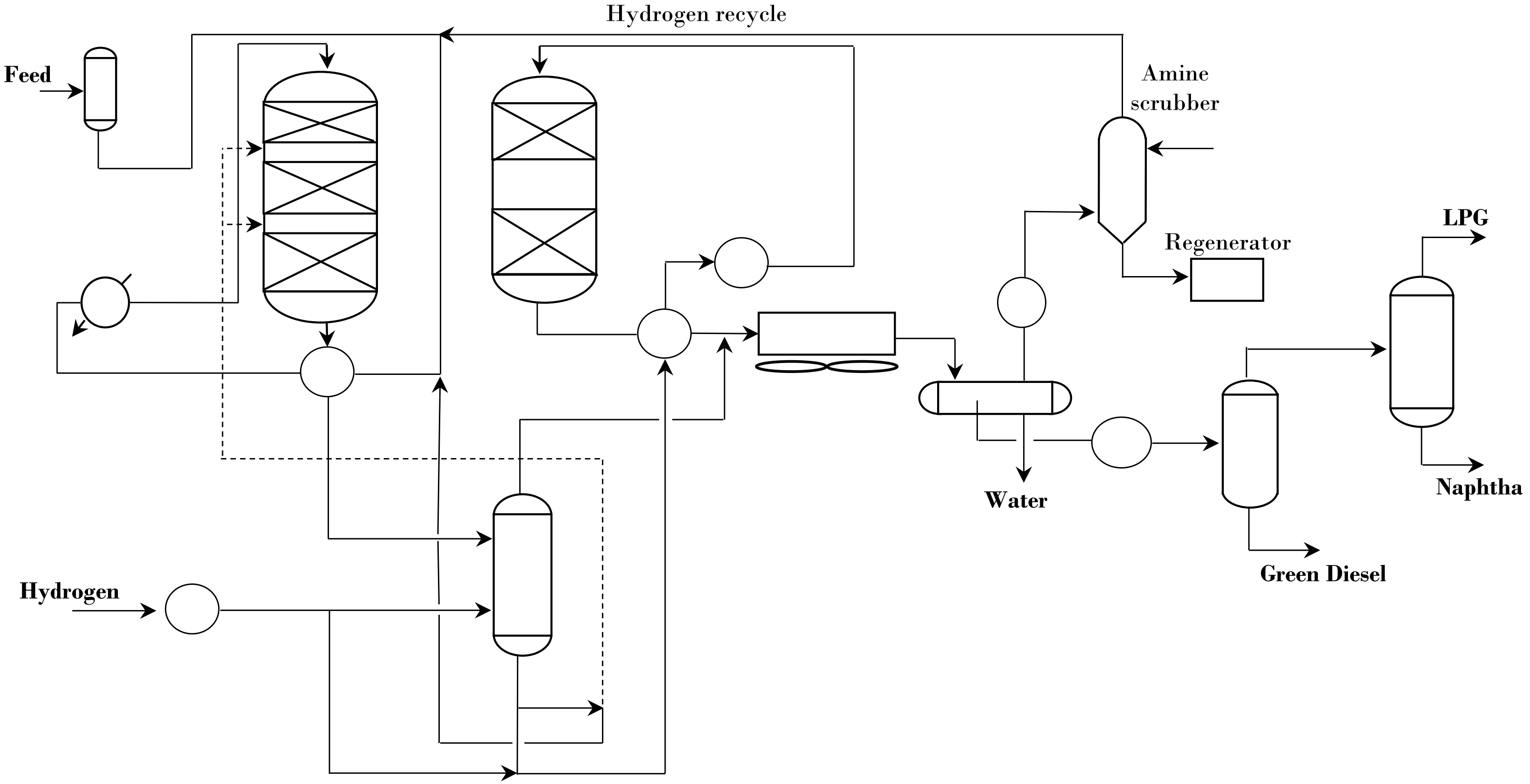


Figure 3.10: Process flow diagram of Ecofining. Adapted from (Bellussi et al.,2016) [10].

In the first reactor, deoxygenation is carried out under temperatures around 300°C and pressures of 3 to 10 MPa, in the presence of a solid sulphided transition bimetallic - i.e. Mo-Ni - catalyst deposited on alumina, in a trickle-bed reactor. Since the reactions occurring are all moderately to heavily exothermic, double quenching via injection of part of the product of this reactor is adopted; a stream of recycled product also mixes with fresh feed, with the same purpose of heat management. Prior to entering the reactor, a furnace preheats the feed-hydrogen mix, which has previously been mixed with the aforementioned reactor products, and with a stream recycled gas from downstream of the reactor section - that is, unreacted hydrogen. This reactor yields n-paraffins and propane, with presence of H_2O, CO and CO_2 in variable proportions, depending on adopted catalyst and operating conditions. A separator downstream of the reactor removes water, carbon oxides and light hydrocarbons - mainly propane - from the main stream. The former stream bypasses the isomerisation reactor and is mixed with its products; the latter stream is mixed with fresh hydrogen and injected in the reactor. If the process is set to maximise diesel fuel production, hydrogen consumption in this stage is very low, and hydrogen serves the main purpose of assuring a pressurised environment. However, hydrocracking reactions will always occur to some extent. This second stage yields the final products: RD, hydrocracking-sourced naphtha and a small amount of light hydrocarbons. This stream is mixed with the bypass stream from the first reactor, cooled down and sent to a fractionator that recovers liquid wastewater and light gases. The light stream is sent to an amine scrubber, in order to separate unreacted hydrogen from the carbon oxides and allow for recycling; the heavy stream undergoes further separation into the different fractions: diesel oil (known with the commercial name of Green Diesel), naphtha and LPG.

Additionally to this plant layout, a pretreatment unit can be optionally featured. This component operates the removal of alkali metals, phosphorus, ash, water and organic impurities such as phospholipids and carotene, which are always present in raw oil and fat feedstocks; it thus enhances feedstock flexibility, allowing to process not only refined oils, but also raw feedstock; the Venice refinery is not yet equipped with such pretreatment unit, whereas the Gela refinery is.

Another difference between the two Italian Ecofining plants is the hydrogen supply unit: in both cases, hydrogen is produced *in loco* by a reformer, which is a naphtha reformer in the case of Venice, and a steam reformer in the case of Gela [10] [83]. The naphtha reformer, inherited from the fossil refinery layout, is said to be the main factor limiting the biorefinery fuel output, and the company is planning to replace it with a new steam reforming unit [83]. This research will investigate an LCA of an Ecofining unit with pre-treatment and steam reformer, thus matching the layout of the Gela plant.

Life Cycle Assessment: model creation

This and the following chapters, which constitute the heart of this study, have the purpose of reporting how the analysis of the GHG impact of cottonseed oil RD was carried out and its implications. Such analysis was implemented following an LCA approach; in its essence, LCA can be defined as "a technique for assessing the potential environmental aspects and potential aspects associated with a product (service), process or activity, by: (I) compiling an inventory of relevant inputs and outputs, (II) evaluating potential environmental impacts associated with those inputs and outputs, (III) interpreting results of the inventory and impact phases in relation to the objectives of the study. The main methodological framework behind the following LCA is the one outlined by the ISO, most relevantly through the international standards ISO 14040 and ISO 14044 [84]. Consequently, the structure at the base of this report follows a division into four phases, as recommended in ISO guidelines: (I) goal and scope definition; (II) Life Cycle Inventory (LCI) analysis; (III) Life Cycle Impact Assessment (LCIA); (IV) interpretation of the results. Phases I and II are covered in this chapter, whereas the next chapter covers phases III and IV.

The LCA is built on a specific case study, in which the production of renewable fuels takes place in an Italian biorefinery, whereas most of the upstream processes, starting from cotton cultivation, takes place in Israel. The case-study analysis is complemented with considerations about the possible extension of its results into a broader context, in line with the objective of the study. As explained in detail throughout the following section, two models have been created, each with its peculiarities in terms of system boundaries and allocation procedures.

4.1 Features of GaBi and LCA models through an example

This work has been carried out using *GaBi 9.2*. This software is designed to assist the user in the development of LCAmodels in two main ways:

- By offering a database structure for the input, management and use of life cycle assessment data.
- By automatically calculating the results once the model is complete.

Every GaBi model is built starting from an existing database, which contains items representing processes. Each GaBi license comes with one or more built-in databases: in this research, it was possible to access the Education database 2020 via direct request to the developers; much effort was put, in some cases, to update or replace existing database information with more recent or more accurate ones from the literature. Any database, in fact, can be expanded and modified. Database information is organised in folders: at the top level, folders collect datasets based on their source; at the lower levels, datasets are organised based on their characteristics, e.g. entire life cycles, manufacturing processes, transport processes; these further split in sub-folders following the same logic.

GaBi works following a strongly modular system, made up of three distinct object types: (I) flows, (II) processes, and (III) plans. Flows are the basis of life-cycle modelling: they serve as representations of actual material or energy flow. Processes roughly correspond to the term "unit process" in ISO 14044 [84, p. 5]: they represent actual processes, defining a set of inputs and outputs (processes exclusively with inputs or outputs can also be defined). Plans are grouping of processes, used to represent a stage or sub-stage of the product system: together, they organise the model hierarchically. Databases organise information in the form of flows, processes and plans; the user can also create new items, either copying and modifying database information, or starting from a blank element.

In order to illustrate in a brief but effective way the features of LCA modelling with GaBi, an example of product system will accompany the following explanation. The example will be based on the life cycle of oat porridge, a popular breakfast in the UK; this particular oat porridge is made with oats and sheep milk - not cow milk, for the sake of the explanation. This life cycle is roughly outlined as follows: milk and oats production, on-farm purchase of the ingredients by a local consumer (supermarkets and other forms of distribution are avoided to keep the example simple), home porridge preparation and, finally, consumption.

We suppose to be interested in evaluating the environmental impact of this oat porridge, in terms of climate change, by measuring its life-cycle GHG emissions; this is the LCA study goal. Fundamental for the LCA study is also the definition of the scope: for example, the analysis may or may not include the production of capital goods, such as farm equipment and infrastructures. The first task for the modeller is to identify system boundaries that are representative and coherent with the study goal and scope; this system could, for example, simply stop at the consumption level, or - more correctly - it could also consider the process of washing mug and cutlery after breakfast. It is equally important to have thorough information about the specific process: is the milk, for example, sold raw or is it subject to on-farm pasteurisation? It is equally important to specify a functional unit for the process system, i.e. a fixed quantity that implies a value for all inputs and outputs: considering the whole system outlined in the example, it could be one ration of

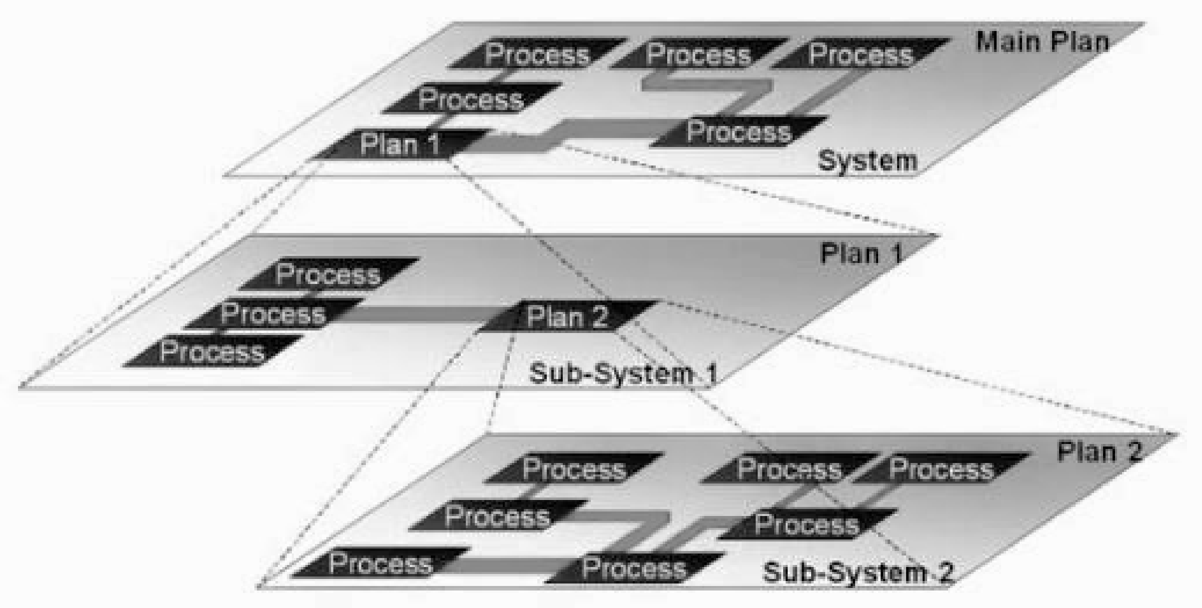


Figure 4.1: A graphic representation of how *GaBi* plans can be nested multiple times.

porridge, where the quantity of each ingredient is set to a determined value like 250 ml of milk and 80 g of oats.

A product system can be modelled in different ways, depending on the required level of disaggregation; let us use oats as an example: all the process steps up to oat production could be aggregated in a single black-box-like unit, with its inputs and outputs (i.e. a process, named *Oat production*) or it could be made up of a plan with the same name, containing processes for different stages of production, for example *Oat cultivation* and *Drying and storage*. Plans can also be nested: *Oat cultivation*, for instance, could be defined as a plan, containing the sub-steps of *Sowing* and *Harvesting* as two distinct processes; nesting can be iterated multiple times, as depicted in figure 4.1.

By focusing on the other ingredient of this unusual porridge, sheep milk, we will now illustrate how a basic balance is computed by the system. The process *Sheep*, as shown in figure 4.2, delivers two useful products: milk and wool. The functional unit of this product system, enclosed within its system boundaries by a red dahsed line, is 1 litre of milk (let's assume a density of 1 $^{kg}/_{L}$ for simplicity): every other flow is thus related to this amount of milk; for example, 0.5 kg of wool may be produced per litre of milk produced. First we will discuss the inputs to the process: supposing that sheep are fed by grazing in local pastures, the only relevant input to the process *Sheep* may be electricity [1] Inputs require the modelling of a supply chain: electricity is produced by the national grid, whose average production mix consumes a certain amount of fossil fuels causing emissions of fossil CO_2. The simplest way to model this is by representing processes as black boxes: this way, there is no need to explicitly model upstream processes such as fossil fuel extraction; the

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[1] The modeller must identify cut-off criteria that allow to restrict the analysis only to what is relevant for the goal and scope of the LCA: for example, antibiotics could be delivered in quantities that make their impact relevant for the study scope, and their supply should thus be carefully modelled. On the other hand, the sporadic use of vaccines may be deemed as irrelevant by the modeller.

emission intensity of this fictitious national grid mix is 2 $^{kg_{CO2eq}}/_{MWh}$, and 1 MWh_{el} is consumed per litre of milk produced. In addition, sheep emit methane during digestion - say 1 $^{kg_{CH4}}/_{L_{milk}}$; in CO2 equivalent, this value is typically considered as 25 times its mass, as methane is a more potent GHG than carbon dioxide. [2] Downstream of *Sheep*, milk and wool undergo distinct processing, i.e. *Milking* and *Wool shaving*: inputs and emissions for each of these processes can unambiguously be attributed to their respective product; *Sheep* itself, on the other hand, delivers *Milk, in udder* and *Wool, on sheep.* The existence of these co-products requires to find a criterium to distribute - allocate - inputs and emissions of farming according to a quantity characterising the flows *Milk, in udder* and *Wool, on sheep*: in the example, the criterium used is simply the mass of each product. The system balance after allocation is shown below in figure 4.2. Note that, besides wool and milk, sheep farming produces also manure, but in the allocation procedure this flow was ignored: this is due to the fact that manure is not generally account for as useful product, but as residue. In fact, when defining a flow, it can enter either of the three following flow types: (I) valuable flows, i.e. flows used in another process; (II) elementary flows, i.e. flows which are taken directly from the environment or released into it; (III) waste flows, i.e. flows that are to be handled in additional process steps, in order to be reduced to elementary flows. Commonly, elementary flows and waste flows like manure are not subject to allocation.

Alternatively to allocation, it is possible to account for co-products by expanding the system boundaries, up to a point where the co-product substitutes an equivalent product of different origin; this method is favoured by ISO guidelines on LCA. Applying system expansion to our example could mean modelling wool processing up to production of a final good - a woollen sweater - which substitutes an equivalent product, e.g. a polyester sweater. All emission up to milk product - 27 kg_{CO2eq} per kg of milk in our example - would be accounted to the milk, but the avoided product system leading to the polyester sweater would provide emission credits to our life cycle: emissions would be saved by replacing a polyester sweater with the woollen sweater. Supposing 10 kg_{CO2eq} arise from processing *Wool, on sheep* into a sweater, and 15 kg_{CO2eq} are accounted in the production of a polyester sweater, the system balance would account 22kg_{CO2eq} to *Milk, in udder*, as shown in figure 4.3. The two major issues when applying system expansion are: (I) knowing with certainty what specific good is going to be replaced; (II) finding a sound equivalence criterium for the replacement.

As the two given examples of co-product accounting through allocation and system expansion point out, the results are often different depending on the methodology used. Here, the same flow of milk is assigned with 18 kg $_{CO2eq}$ with mass allocation and 22 kg_{CO2eq} using system expansion.

A variation of this example can be used to explain another feature that GaBi conveniently allows to model. Suppose that sheep are fed with oats instead of grass; suppose that these oats are harvested and processed locally, and that these same oats are bought

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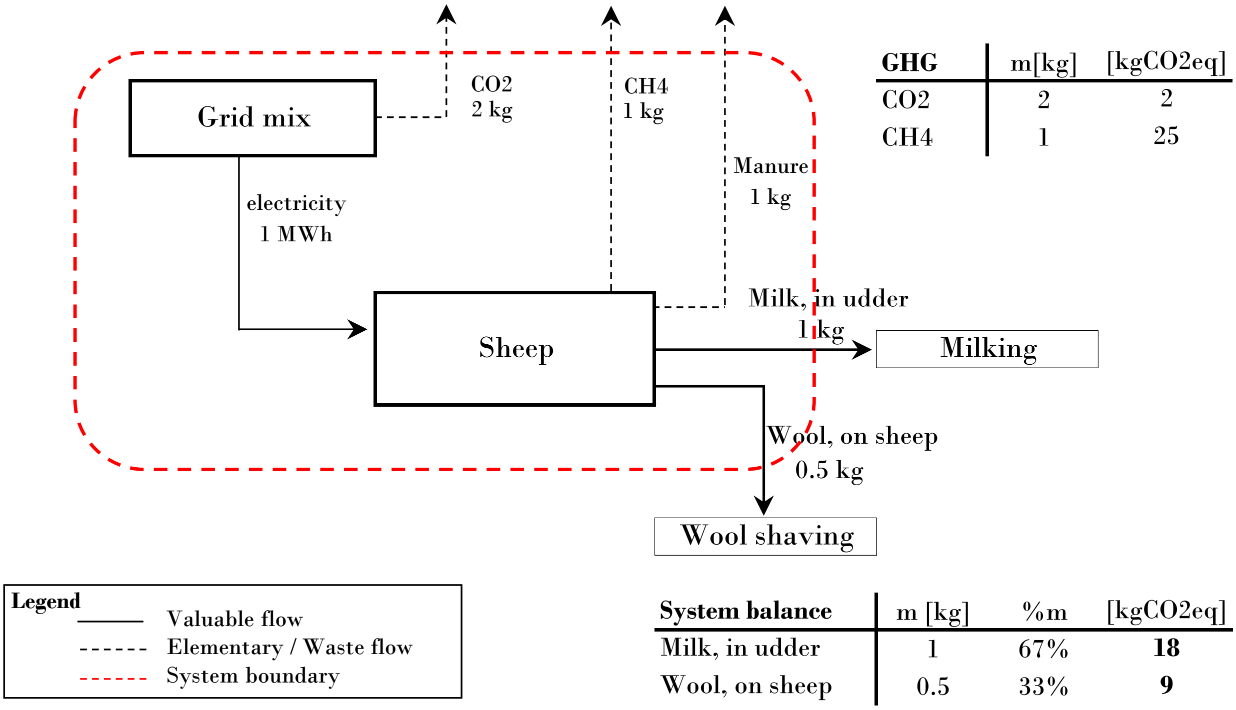
[2] The emission factor of methane is commonly considered $^{kg_{CO2eq}}/_{kWh}$, which corresponds to a time horizon of 100 years; a shorter time horizons would result in higher emission factors, as the GHG effect of this gas reduces with time thanks to its gradual decomposition.



GHG	m[kg]	[kgCO2eq]
CO2	2	2
CH4	1	25

System balance	m [kg]	%m	[kgCO2eq]
Milk, in udder	1	67%	**18**
Wool, on sheep	0.5	33%	**9**

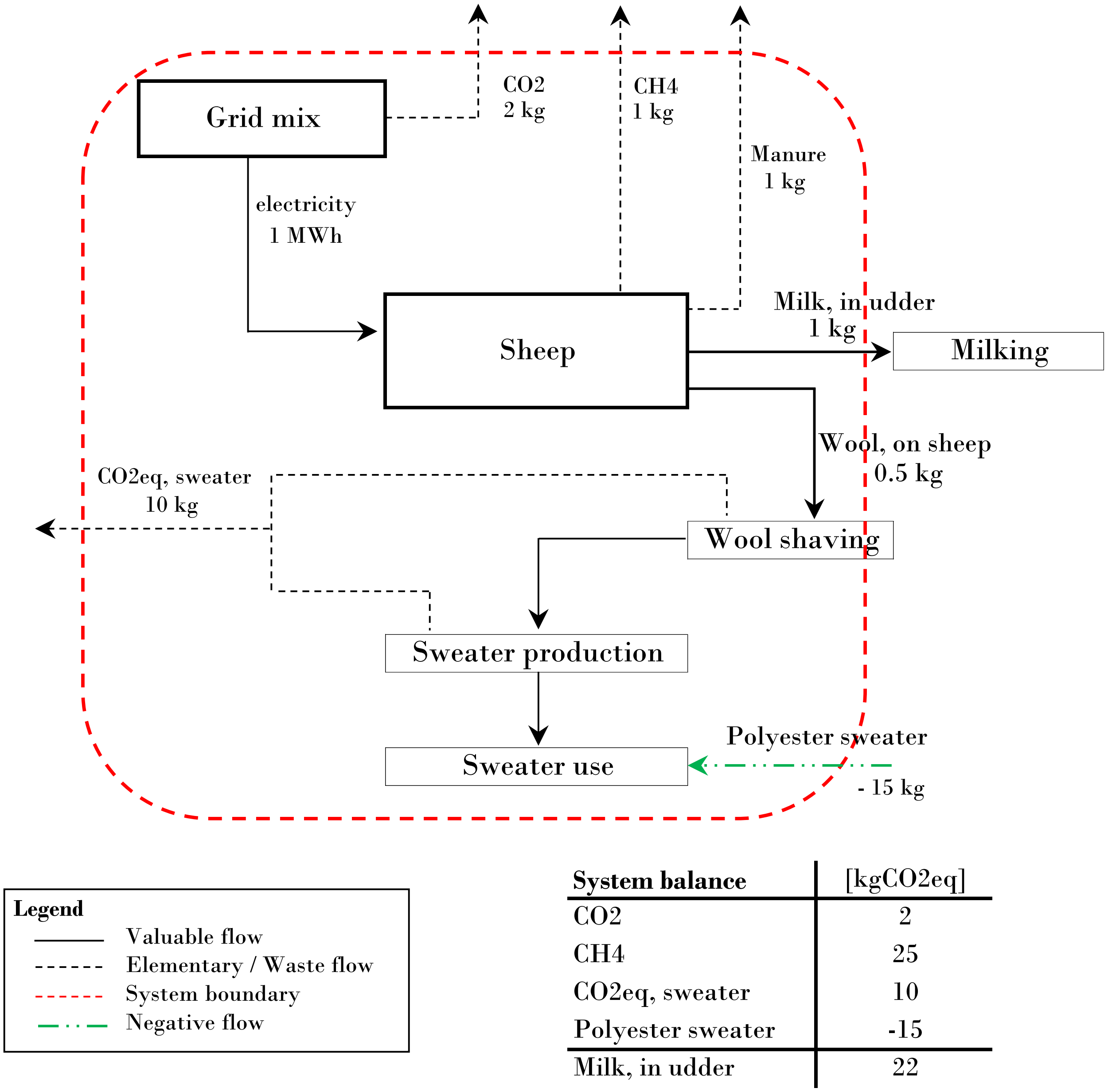
Figure 4.2: System balance of sheep milk production in the current example, using allocation.

by the local customer to make porridge. In this case, the process or plan of *Oat production* can be modelled in two distinct instances and employed in the two different points of the life cycle. In fact, processes as defined in the database are distinguished from process instances; process instances are independent calls of a database element, and can be different in their parameter definition and/or in their allocation, if present. In this case, for instance, there may be a difference in the parameter *Storage moisture* between the two oats instances.

After setting up the model, GaBi allows to automatically calculate the system balance. Results analysis can be carried out, in GaBi, through sensitivity analysis and Monte Carlo simulations, relying on the parameters set by the modeller, as it will be discussed in section 5.2.

4.2 Goal and scope definition

This LCA concerns the production of renewable diesel (RD) through hydroconversion of cottonseed oil. Its first objective is the evaluation of GHG emission reductions of this biofuel in comparison with the fossil alternative, i.e. fossil diesel oil, in order to evaluate



System balance	[kgCO2eq]
CO2	2
CH4	25
CO2eq, sweater	10
Polyester sweater	-15
Milk, in udder	22

Figure 4.3: Product system configuration of sheep milk production in the current example, using system expansion for the co-product wool.

this fuel in the context of the latest EU regulations and verify its compliance therewith. The second objective is the comparison of cottonseed oil with other competing feedstocks for the production of RD, in terms of life-cycle GHG emissions.

The scope of the study focuses on a product system aimed at modelling the physical system that leads to the biorefining of cottonseed oil starting, in principle, from the cultivation of cotton, with the function of producing renewable transportation fuels, and, most notably, diesel fuel. The initial stages of this process are considered as being located in Israel, whereas the biorefining process unit is considered to be located in Italy. In this country, cottonseed currently undergoes a different use: after ginning, in fact, cottonseed is sold as it is to local dairy farmers with an auction mechanism; such a use is common to many contexts worldwide; this research thus adapts well to all contexts where cottonseed is employed without further processing as feed in the cattle industry, which is by far its main application.

The primary reason for carrying out this study is linked to the most recent developments in EU regulations, primarily through its last directive RED II, which are forcing many RD producers to rapidly find suitable substitutes to palm oil, considered unsustainable and thus destined to be banned as biofuel feedstock by 2030, for the compliance with EU national targets. Cottonseed oil has not been interested by the same level of research that other oils and fats have been exposed to over the years: the aim is thus to cover this research gap both in the framework of EU regulations and in the framework of the most commonly approach to LCA, i.e. following ISO principles.

This research can be useful to policy-makers and, most importantly, to actors in the EU biorefining industry, which may consider cottonseed oil as an addition to their portfolio, especially for a medium-term replacement of palm oil.

The analysis is carried out by means of two parallel models, where different logics are applied to the product system. In the first model, or EU Compliance Scenario (EUC) model, the system is adapted to follow indications as indicated by RED II for the calculation of GHG emission reductions. In the second model, or IS model, the system follows ISO norms ISO14040, ISO14044, ISO14046, which are the main reference used in LCA studies worldwide. Applied to the present study, these methodologies translate into differences in the treatment of co-products on two levels, i.e. at the cotton gin output level and at the oil extraction output level, as explained further in the section. An additional goal of the research is thus to compare the impact of different methodologies on the final results.

The main methodological differences between these two methodologies are highlighted in table 4.1.

Table 4.1: Main differences between the two LCA methodologies featured in this work.

Methodology	**Agricultural residues treatment**	**Co-products treatment**
EU RED II	by-products (mandatory)	energy allocation (mandatory)
ISO	co- or by-products (discretionary)	system expansion (preferred)

As shown in table 4.1, the ISO methodology admits system expansion and also different allocation methods; in this respect, system expansion is to be preferred: moreover, in case it is deemed more appropriate to adopt allocation, it is suggested to base it on physical properties rather than on other quantities, such as market price. The EU, on the other hand, requires to strictly apply energy allocation. [3] In its guidelines, system expansion is not allowed at all.

It is now appropriate to discuss the distinct characteristics associated with each of the two methods at the base of the EUC and the IS models. The methodological differences between EU and ISO guidelines translate in fact into the different strengths and weaknesses shown in table 4.2.

Table 4.2: Methodological differences, strengths and weaknesses of the two methodologies implemented in the current research, following ISO norms and EU REDII rules respectively.

Method	**Features**	**Strengths**	**Weaknesses**
ISO	- System expansions favoured over allocation - Co- and by-product treatment is not bindingly set	- more flexible modelling - More widespread	- It can be used tendentiously
EU RED II	- Energy allocation mandatory - Agricultural residues defined as by-products	- Necessary for checking compliance with EU RES targets - No room for misleading interpretations	- It may omit important impacts

By admitting only energy allocation, the EU methodology is clearly more rigid: on the one hand, it eliminates the risk of murky results that a tendentious or naive application of ISO guidelines may allow; on the other hand, it may omit important impacts. ISO guidelines, in fact, may lead to much different results depending on the modeller's decisions: one clear example of this is applying system expansion for co-produced fuels (in our case, these would be renewable naphtha and renewable LPG) which may alter the results to a great extent and can even lead to a negative Global Warming Intensity (GWI) of the main product [85] [86].

The EUC model serves mainly a regulatory purpose, whereas the International Standard (IS) model is better suited for a more holistic capture of all implications of the process, and for comparing this with other feedstocks; system expansion, in fact, capture effects such as ILUC and pressure on competing markets, which are not considered in the EUC model. Additionally to these two main models, a number of scenarios will be covered in section 5.2, all using the IS model as basis.

Figure 4.4 shows a representation of the proposed product system for Israeli cottonseed, highlighting the level of disaggregation implemented and the system boundaries for each of the two models. Here, it is possible to appreciate the differences between the EUC and the IS model in terms of system boundaries; for clarity purposes, it shows only the main

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[3] The last reviews of the EU methodology as required by RED II actually feature an exception to this rule: in case of cogeneration of heat and power, exergy allocation is required; this, however, does not concern the present research.

intermediates and outputs.

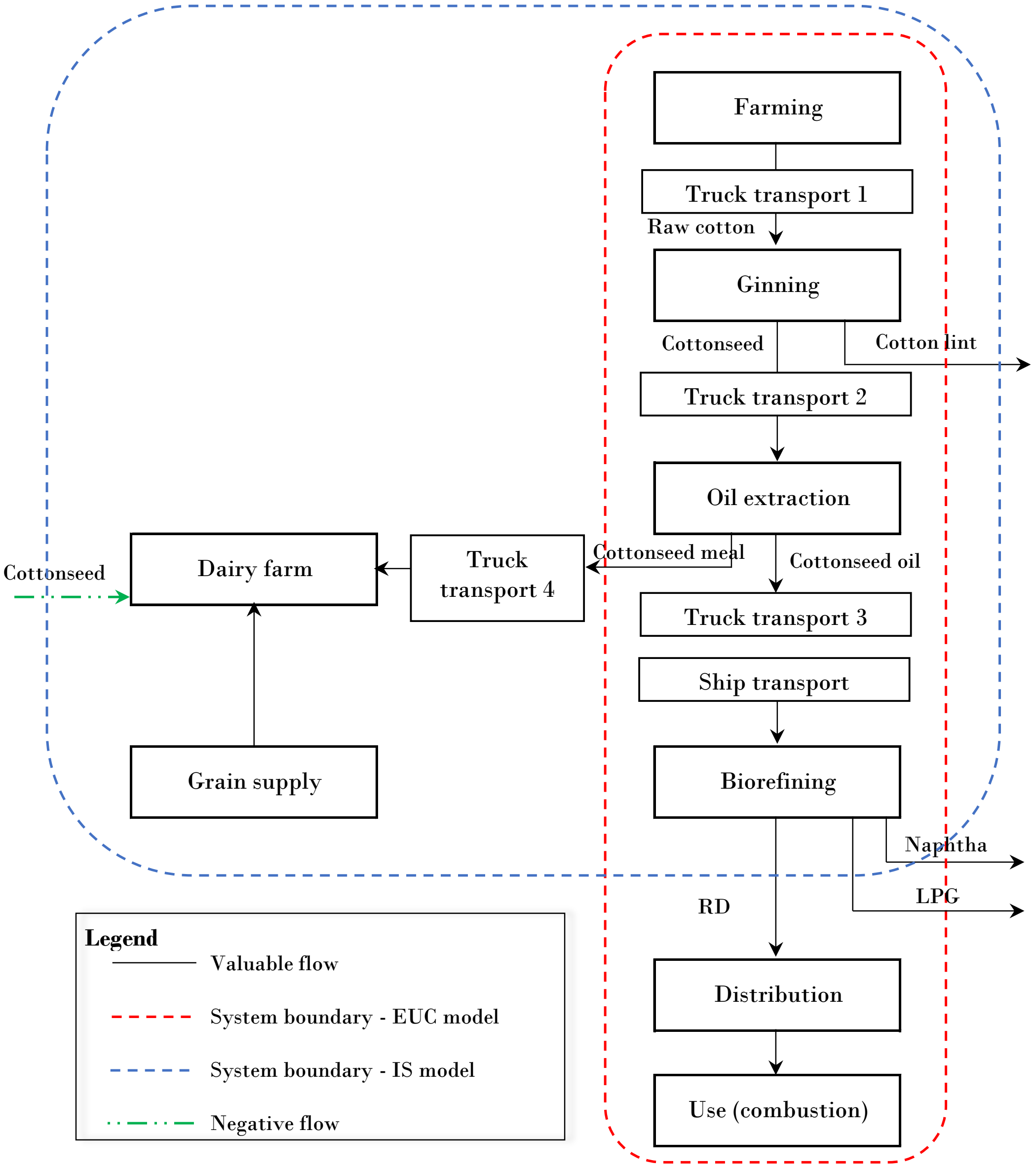


Figure 4.4: Representation of the product system analysed, showing the system boundaries according to the two different models: EUC (red dashes) and IS (blue dashes). Waste flows and emissions are omitted.

In the case of the EUC model, a "well-to-wheel" LCA is carried out, where system boundaries end after diesel fuel distribution - and also combustion in the engine, which, given the fact that biofuels are made of renewable biogenic carbon, does not account for any increase in GWI. The inclusion of these two life-cycle step is dictated by EU regulations; distribution will be modelled after the default distribution emissions provided by EU regulations [7]. In the case of the IS model, system boundaries characterise it as a "cradle-to-gate" LCA, i.e. ending at the biorefinery output level. This choice allows to compare cottonseed with other competing feedstocks without including fuel distribution, which is a highly uncertain process and also generally responsible for low to negligible levels of emissions.

The main process units considered are: cultivation, ginning, oil extraction, biorefining; in-between each step, a transportation unit process is modelled as well, coherently with the mode of transport of the corresponding physical system. This disaggregation is in line with the actual material processing underlying the model: it allows for the univocal identification of co-products, while allowing a level of detail in line with the goal of the study.

As shown in figure 4.4, the two sets of guidelines have many diverging implications on the flows linked to these process units. EU regulations impose that, first of all, whole cottonseed at the gin output level be counted as having no environmental impact; in addition, the approach to the two multi-product systems downstream of the ginning facility must be uniquely by allocation through energy content (LHV). In contrast to this, ISO guidelines prioritise system expansion over allocation: for this reason, at the level of oil extraction outputs, system expansion instead of energy allocation is adopted to account for cottonseed meal being delivered to an Israeli dairy farm. Here, cottonseed meal is used as cattle feed, substituting a corresponding amount of whole cottonseed. Such system expansion requires the modelling of a grain supply element: although not explicitly shown in the figure, this unit contains the model of the whole grain supply chain up to delivery at the dairy farm, which depends on the specific grain under consideration; many different grains, in fact, will be considered.

Another relevant difference is the possible presence of allocation at the level of gin outputs, where EU regulations impose to treat whole cottonseed as a residue with no environmental impact, while ISO guidelines allow to account for cottonseed as a co-product of cotton lint, thus assigning an environmental impact thereto. In fact, the treatment of items like agricultural residues as co-products or by-products has never reached universal consensus, as witnessed by the large amount of LCA literature following ISO guidelines which adopts either the first approach [87][88][14], or the second approach [89][90][91]. A strength of the IS model is the contextual presence of a positive and a negative flow of whole cottonseed. This feature is given by the specific final use of wholeseed as cattle feed in the current business-as-usual case, which allows to model the system in such a way. The two flows are characterised by the same magnitude in the system: as a consequence, adopting the former or the latter approach in matter of treatment of agricultural residues does not have any consequence on the model net results. This feature, which is explained more in depth in section 4.3.1, makes the model more flexible than the usual agricultural LCA, and capable of being compared with other studies regardless of the approach they adopt.

Finally, at the level of the biorefinery outputs, both models adopt energy allocation. In principle, ISO guidelines would prefer the modeller to adopt system expansion: however, as already pointed out, this approach has been proven to lead to misleading results when applied to multiple fuel outputs. For this reason, energy allocation is chosen.

As pointed out in the introduction, there is a substantial lack of research for cottonseed biofuels and RD in particular. Additionally to this issue, the LCA development had to face a discrepancy between industrial data on RD production and existing literature, and in particular about naphtha yield, which is thoroughly explained in the related section about the biorefinery model 4.3.4. Another issue is linked with the estimation of emissions related to indirect land use change: these have here to refer to cereal grains for RD production, whereas published studies invariably assign these emissions as coefficients related to the unit of biodiesel or bioethanol [92] [93] [94] [95].

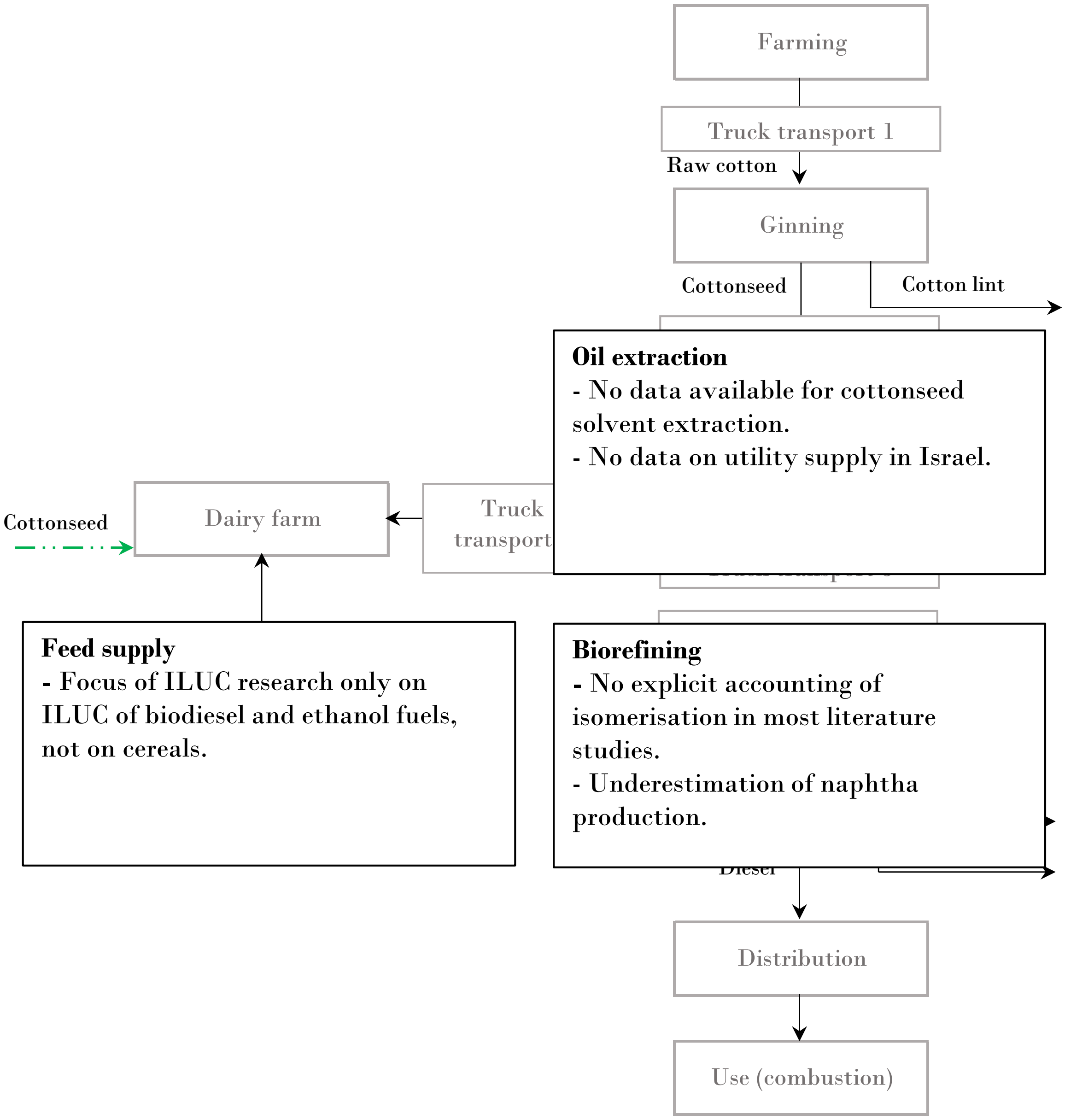
The major issues in the study development are collected in figure 4.5. Additionally to these, other data gaps had to be faced with the development of model modules from their foundations, as in the case of the Israeli electricity grid and the fertiliser supply chain.

The adopted functional unit is 1 MJ of energy content (LHV) of RD produced, in both models. Data for the elaboration of the LCA have been calculated or estimated relying either on first-hand data or on secondary sources; the primary source of secondary data were peer-reviewed scientific articles, which have been used together with database information sourced from the GaBi database - such as from the GaBi refinery model for what concerns the production of fossil fuels. further information about the data collection procedure for each process unit and flow are reported in the following section. The carbon equivalent of each flow includes the production of all material and energy inputs necessary for its fabrication and its supply to the point of utilisation, as well as any emission related to its production and supply; the discounted carbon equivalents deriving from fixed capital goods are not included in the analysis. All inputs considered relevant for the study, matching the degree of precision required, have been included in the assessment; the cut-off criteria, in case some minor input has been excluded, are outlined in the corresponding inventory sections. In this respect, it is relevant to state that, coherently with common practice, inputs of services such as cleaning, accounting, marketing - research and development activities and overhead are excluded from the system. Being based on a case study, the present research is not intended to be universally representative for such a process of RD production from cottonseed oil. This said, the results may be adapted to other contexts where cottonseed is used whole as cattle feed.

4.3 Life Cycle Inventory

4.3.1 Cotton cultivation and ginning

In principle, cotton cultivation and ginning are to be included in the LCI analysis. Cultivation is the most important contribution in most agricultural LCA, especially given the high emissions linked to fertilisers; ginning impacts, on the other hand, are mainly related to energy inputs in the form of electricity and natural gas for drying.



Oil extraction
- No data available for cottonseed solvent extraction.
- No data on utility supply in Israel.

Feed supply
- Focus of ILUC research only on ILUC of biodiesel and ethanol fuels, not on cereals.

Biorefining
- No explicit accounting of isomerisation in most literature studies.
- Underestimation of naphtha production.

Figure 4.5: Representation of the product system, highlighting the major issues involved in its modelling.

Notwithstanding, the present LCA allows to exclude these processes from the computation;

more precisely, the analysis can neglect any impact coming from processes upstream of cottonseed output at the cotton gin. This is possible thanks to the peculiarities of the two models under consideration, which, in different ways, do not feature cotton cultivation as having any impact on the life cycle of cottonseed oil HVO.

In the EUC model, the product system features only energy allocation. Most importantly, EU indications fit cottonseed into the definition of residue, to which no environmental burden is to be assigned; for this reason, all processes up to cottonseed do not play any role in the final results, and can be neglected.

In the IS model, on the other hand, it may be possible to exclude cotton cultivation for the same reason, in case cottonseed is defined as a by-product without any burden. In fact, ISO guidelines do not set rigid rules with regards to by- and co-product treatment, which means that, in principle, cottonseed may be assigned a burden together with cotton lint; with such approach, cultivation would have to be included in the analysis. Even in this case, however, the IS model would not require to study cotton cultivation for computing the results, thanks to the system expansion at the dairy farm: in fact, cottonseed meal replaces whole cottonseed as animal feed, together with the supplement of other energy feeds; this allows the net accounting of whole cottonseed in the model to be zero, with a positive flow and a negative flow of the same magnitude downstream of it. The only difference that impacts on the final results is constituted by the different supply chain, which includes different transport steps - the positive flow is transported from the gin to the extraction plant, whereas the negative flow is transported from the gin to the dairy farm; this, of course, is accounted for in the model, as explained later in the relative sections.

Besides cotton lint and cottonseed, the process of ginning also delivers a flow of cotton trash, in the proportion of about 10 to 12% by weight [96] [97]. Its presence does not influence the outcome of any model, since it fits into the definition of a waste flow.

4.3.2 Cottonseed oil extraction

After being separated from the lint, whole cottonseed is transported to an oil extraction facility. The goal of this process is to fraction the seed into its constituents: hulls and linters, meal, oil. It was not possible to obtain and analyse samples of Israeli cottonseed, therefore seed composition was estimated using literature data. After a thorough literature review, it emerged that such composition is subject to remarkable variability: including a presence of 4 to 7 % trash, lean meal constitutes 42 to 45.5% of the seed by dry weight, hulls 25 to 27%, linters 8 to 10%, and oil content is in the range of 15 to 18 % [98] [99] [100] [101]; other studies cite a value of 20% [102], with references to oil contents as high as 25 % [103] [104]. Edwards et al. state an oil content of 20%, with a range of variation from 10 to 28% [105]. In the present study, a somewhat conservative oil content of 18% as base case is adopted; the shares of other constituents were also assumed based on literature data, as shown in table 4.3; the reported values are net of the trash content of 5 %, also determined based on its prevalence in the literature.

Table 4.3: Cottonseed composition adopted in the present study baseline (dry weight).

Cottonseed composition		
Oil	% DM	18
Lean meal	% DM	46
Hulls	% DM	27
Linters	% DM	9

This part of the process could not be modelled starting from an actual production process, since cottonseed is currently sold whole to the local dairy industry. Nonetheless, Israel hosts many oil extraction facilities and vegetable oil refineries, as witnessed by its imports of oilseeds: in 2019, imported rapeseed in the country amounted to 161 kton, and imported sunflower seed to 216 kton [106]. It is thus possible, in principle, to employ an existing facility to extract oil from cottonseed, adapting its parameters to the peculiarities of cottonseed. As the next paragraphs will illustrate, in fact, seed oil processing does not differ much depending on the feedstock.

The typical process to extract cottonseed oil starts with a screening to remove dust and other residues. It is common practice, after such screening, to proceed removing linters from the cottonseed surface, then remove seed hulls prior to pressing [3]; this way, linters become a valuable marketable product. In the present study, the process features a direct dehulling of lintered seed, so that linters become a component of the meal product. Such a process is simpler and thus compatible with existing edible oil extraction facilities, since delintering is proper of cottonseed processing only [107]. The implications of this choice will be analysed further on in the section.

Once dehulled, the seed can undergo oil extraction either by simple pressing, or by pressing and subsequent solvent extraction; the latter process is very common in the vegetable oil industry, and it guarantees higher oil yield, especially when treating seeds with oil content lower than 30% like cottonseed [101]; for these reasons, it is the one adopted as base case in the present study. The solvent used is normally n-alkane - most commonly n-hexane; some losses of solvent to air due to incomplete recovery are always to be expected.

When solvent-extracted, oil sources with relatively low oil content like cottonseed and soybean are normally processed via direct solvent extraction, i.e. without a mechanical pre-pressing [101]; such a process is thus made up of the following steps: screening, cracking and dehulling, pre-heating, cooking, pressing, solvent extraction. In general, raw vegetable oil for human consumption is subsequently refined through a series of processes - typically degumming, neutralisation, bleaching, deodorisation - prior to distribution; in our study, it was assumed that raw cottonseed oil would not undergo a complete refining, but would be treated in the pretreatment unit at the biorefinery.

Data for each process step were gathered from the literature. Since no reliable data can be found with regards to cottonseed oil extraction - except for a simple pressing of cottonseed oil operated in rural Brazil [2] - the study focused on data from soybean oil extraction facilities. This approach is supported by the fact that these two oil sources share similar oil content and therefore very similar process layouts, unlike rapeseed and other oilseeds. Nonetheless, it requires adaptation of the data to better fit the properties of cottonseed processing. It was decided to proceed by collecting data from many different oil extraction facilities, while keeping soybean as reference for the modelling of cottonseed processing as well.

Data gathering focused on sources which allow for a disaggregation of energy inputs for drying, which is not included in the present analysis since cottonseed enters the facility at a moisture level of 7% if directly from the gin, and hardly above the suggested threshold of 10% [78] [101].

As shown in table 4.4, two different soybean datasets have been analysed: the data in (JRC,2017) and the data in (Reuters,1998). The latter study allows for the finest level of disaggregation, thus offering a clear distinction of energy need by process step[78]. However, the selected study was published in 1998, which means that consumptions may not reflect the state of the art; it was deemed as necessary to compare the aforementioned studies with data from other oil extraction facilities, as reported in table 4.4: five LCIs have been compared, treating soybean (2), rapeseed (2) and sunflower (1). Input and output data have been uniformed to reflect a unit mass of 1 kg of as-received oilseed feed.

Table 4.4: Literature data of total material and energy inputs for oil extraction of soybean, rapeseed and sunflowerseed.

	(Reuters,1998)		**(JRC,2017)**		**(Schmidt,2007)**
Feedstock	**Soy**	**Soy**	**Sunflower**	**Rape**	**Rape**
Steam $[MJ/kg_{AR}]$	0.90	0.58	0.55	0.67	0.66
Steam $[MJ/kg_{oil}]$	5.4	3.0	1.3	1.6	1.6
Electricity $[kWh/kg_{AR}]$	0.056	0.029	0.050	0.042	0.049
Electricity $[kWh/kg_{oil}]$	0.41	0.15	0.12	0.10	0.12
Hexane $[g/kg_{AR}]$	2	0.60	0.60	0.80	0.30
Oil content $\%dry$	21.6%	21.6%	48%	46%	46%
Source	[78]		[18]		[108]

The first modelling step was the choice of an extraction yield reflecting the efficiency of the process. In this respect, for solvent-extracted rapeseed meal, oil content in the meal is said to normally vary between 0.7% and 1.5% [101]; this would translate into an extraction yield of 93 to 97% for cottonseed. The older soybean extraction facility featured in (Reuters,1998) has a yield of 95%, whereas the modern soybean plant in (JRC,2017), based on the reported values, presents a unitary yield, with all other extraction processes featured in this source close to such value; similarly, the rapeseed plant in (Schmidt,2007) features a yield of 98.5%.

Based on these observations, the baseline yield is here set at the conservative value of

95%; this value takes into account the possibility of a yield reduction due to the absence of seed delintering prior to dehulling. The value is then supposed to be possibly as low as 90% and as high as 99%. 90% is thus taken as a minimum representing a solvent-extraction plant with high loss due to absence of delintering, whereas 99% represents a state-of-the-art extraction plant with a delintering process prior to dehulling.

By knowing oil yield and incoming moisture, it is immediate to determine the baseline values of mass of seed in both dry and as-received terms, and also oil leakage into the meal. Setting a dry seed mass also allows to determine, under the assumption of no mass losses during the process, the outputs of lean meal and lintered hulls (hulls+linters) from the values in table 4.3.

In the model, oil yield and oil leakage into cottonseed meal are parametrised according to equations 4.1a and 4.1b: these simple formulas will allow to relate the oil yield η_{oil} to the oil net output and the oil content in the meal. Here, oil_{dry} represents the oil content in the dry seed - 18% under baseline conditions. This parametrisation is also useful to assess the meal LHV in relation to its content in oil, and to its nutritional properties, aspects which will be discussed further on in the section.

$$Oil_{output} = Feed_{dry} * oil_{dry} * \eta_{oil} \tag{4.1a}$$

$$Oil_{leaked} = Feed_{dry} * oil_{dry} * (1 - \eta_{oil}) \tag{4.1b}$$

Focusing now on utilities consumption, it can be noticed that, at a first glance, the older dataset for soy from (Reuters,1998) features much higher utility consumptions per unit of oil yield compared to the other datasets. By comparing soybean datasets with sources for different feedstocks, it emerges that while steam consumption appears to have a tighter link with incoming as-received mass, electricity seems to be more tightly linked with to the unit of net oil output. This was the starting point for the estimation of a plausible value for cottonseed utilities, which are here unavoidably subject to large uncertainties.

In matter of steam consumption, it can be noticed that, if we exclude the older dataset from (Reuters,1998), values are rather similar per unit mass of feed, centred around 0.61 $^{MJ}/_{kg}$, with a standard deviation of 10% (versus 1.85±40% $^{MJ}/_{kg}$ per unit of net oil output): as a consequence, feedstocks with lower oil content will bring about higher specific steam consumption per unit of extracted oil. To the author's judgement, the discrepancy in steam consumption of older data may find explanation especially in: (I) a less efficient steam-meal heat exchange, linked to the older design of the desolventiser-toaster; (II) the lack of heat recovery. This second factor, in particular, is reported to be of key importance in modern oil extraction plants [101]. For these reasons, the steam demand of the soybean crushing facility in (JRC,2017) is adopted as base case, i.e. 0.58 $^{MJ}/_{kg_{AR}}$ of cottonseed. This value will be tested in a parameter variation, varying it of ±25%, given the restricted range of variability observed in steam consumption from table 4.4. The assumption introduced in the modelling of cottonseed oil extraction is thus that its steam consumption is proportional to incoming wet mass of seeds. This assumption seems to be confirmed by comparative literature on soybean and rapeseed oil extraction as well [109]. The baseline value for steam consumption was set at the same level of the soybean facility in (JRC,2017), i.e. 0.582 $^{MJ}/_{kg_{AR}}$ of seed. In terms of specific consumption over dry mass or oil yield, both the lower oil content and the lower oil extraction yield of the modelled cottonseed plant make consumption higher than the one featured in the soybean facility.

The data on specific electricity consumption features a higher variability between soy and other feedstocks in terms of as-received feed, and soy from (JRC,2017) may be interpreted as an outlier with respect to the other values; on the other hand, the consumptions in analysis seem more closely related when specific to the net oil output: the data are centred on 0.12 $^{kWh}/_{kg}$ of net oil output, with a standard deviation of ±17%, compared to a higher range of variation in terms of as-received feed (0.043±23% $^{kWh}/_{kg}$). Again, cottonseed specific electricity consumption is assumed at the level of the one for soybean in (JRC,2017), i.e. 0.15 $^{kWh}/_{kg}$, this time adopting the oil-specific value. Starting from this assumption, a difference is assumed to rise from the dehulling process, which is expected to be more energy-intensive for cottonseed, per unit of processed mass: this would be due to the harder hulls of cottonseed compared to soybean.

In order to account for the difference in dehuller electricity consumption, this research had to rely on the dataset from (Reuters,1998), which is the only study found to report electricity use at this level of disaggregation; in this research, the fraction of consumption devoted to dehulling constitutes 14.1% of total electricity consumption. By assuming that such proportion would hold true also for a more modern plant such as the one featured in (JRC,2017), it was possible to disaggregate this consumption in the newer process as in (JRC,2017). Then, the processable load was estimated based on a commercial datasheet, where it is reported between 33 and 46% [110], meaning that such dehulling machine, when processing cottonseed, is able to process less than half the mass flow rate of soybean. An average of 39.5% is thus here utilised as baseline, and the extremes will be tested in a parameter variation. Equation 4.2 shows how this calculation is computed, with values for the baseline case.

$$el_{cott} = el_{tot,soy} * [\frac{el_{dehull,soy}}{el_{tot,soy}} * (r_{dehull})^{-1} + (1 - \frac{el_{dehull,soy}}{el_{tot,soy}})] \tag{4.2}$$

el_{cott}	Cottonseed total electricity consumption	$[^{kWh}/_{kg_{oil}}]$	0.182
el_{soy}	Soybean total electricity consumption	$[^{kWh}/_{kg_{oil}}]$	0.150
r_{dehull}	Load ratio at dehuller	$[^{kg_{cott}}/_{kg_{soy}}]$	0.395
$(^{el_{dehull,soy}}/_{el_{tot,soy}})$	Portion of electricity for dehulling	[%]	14.1

Thus, the resizing of dehulling consumption causes an increment of about 20% in electricity consumption in our model baseline case.

It is now worthwhile to provide some more context for the process of dehulling. Dedicated machines, commonly called dehullers, exist in different designs, some of which are able to process a spectrum of different materials such as soybean, sunflower and cottonseed: a corrugated roller mill dehulling machine, as the one used in (Reuters,1998), is capable of processing both seeds [110]; in this research, specific electricity consumption of this kind of machines was re-scaled according to the mass of material being processed. As a final remark on dehulling lintered seed , it is to be noted that the process of dehulling lintered seed (i.e. seed that did not undergo delintering before dehulling, as in our analysis) may further reduce the actual processing rate, or it may even not be possible at all in some machines; additionally, it may reduce the final oil yield, as linters may hinder the separation of seed meat and hulls, causing some meat to be lost within the hull flow. These effects are subject to large uncertainty, as no specific data could be gathered. These detrimental consequences are somewhat implied in the parameter of oil yield, which, at 95%, is some percentage points lower than all other observed values.

Note that both the term r_{dehull} and the utility consumption terms are determined on the basis of as-received mass. They are thus function of the moisture content of incoming cottonseed: the higher the seed moisture, the worst the environmental performance of this unit process. This aspect has been taken into account in the model by parametrising seed moisture in their definitions. To determine this moisture value, cottonseed can be considered to exit the cotton gin at 7% moisture, which is the upper limit of moisture for proper cotton cleaning and handling [96]. During transportation and storage, this moisture level tends to adjust depending on environmental conditions: at 50% relative humidity and 21°C, the seed equilibrium moisture is about 10%; if relative humidity reaches 70%, this value increases to 12% [96]. Being Israel a hot and dry country and being indoor storage preferable over outdoor storage, we assume a moisture content of 10%; this value will also be subject to testing, between the values of 8 and 12%.

Finally, note that it is possible, in principle, to use seed hulls on the spot to produce thermal energy via combustion: this would make heat demand of the oil extraction facility self-sustaining [111]; in this research, a conservative approach was adopted: cottonseed hulls are thus collected and mixed into the seed meal.

Hexane leakages differ by one order of magnitude between (Reuters,1998) and all the other sources: such a difference appears to be caused by technological progress, as, in both cases, hexane leakage from the two rapeseed plants is in line with the more recent data on soybean: for this reason, the value from [18] was adopted; this approach is supported by the similarity of the hexane recovery section of these plants, regardless of the specific feedstock.

The literature review also provides additional data from an Indian cottonseed oil extraction facility, published but not peer-reviewed, which set the electricity consumption at 0.12 $^{kWh}/_{kg_{seed}}$ and steam consumption at 0.68 $^{MJ}/_{kg_{seed}}$ (knowing that a gas boiler is in use, and adding the assumption of 80% boiler efficiency). This electricity consumption is not in line with data for other feedstocks as in 4.4, whereas steam consumption is well in line with what demanded in the other cases, per unit of feed; this last fact strengthens the assumption, knowing that the highest impact is given by steam demand rather than the electricity demand.

Table 4.6 collects all the inventory data for this process step, which have been discussed so far. Quantities are reported on the base of 1 kg of oil net yield. The process is modelled as a single unit, as in figure 4.6, which shows flows per unit of oil output; the reported value for electricity consumption contains also the contribution from dehulling.

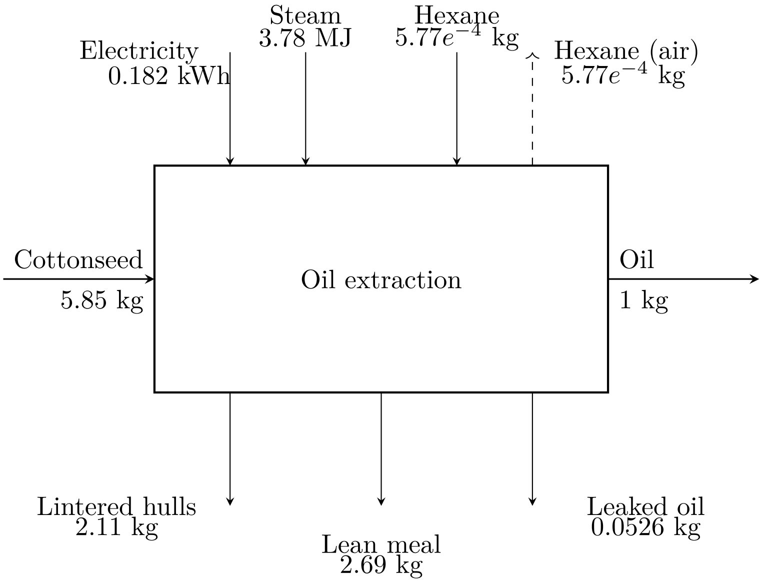


Figure 4.6: The oil extraction unit process with its inventory flows, relative to 1 kg of net oil output. Material flows reported as dry matter; their totals may not add due to rounding.

Downstream of the process, the flows of cottonseed hulls (lintered) and cottonseed lean meal are mixed, together with the leaked oil content, forming a single stream of cottonseed meal. Waste, as well as waste treatment are neglected; as in previous process steps, inputs of lubricant and chemicals for cleaning and maintenance are omitted as well. Steam is assumed to be produced from a natural gas boiler at 85% efficiency, using GaBi database as data source. Electricity supply is modelled after the Israeli electricity grid average inputs, as discussed in section 4.3.6. Hexane supply is modelled after data available in the European Commission JRC database, the same database used for the calculations of the typical and default GHG emission values in the 2016 RED II draft [112].

The calculation of the LHV for each output, necessary for energy allocation as in model EUC, is based on the properties of each component; in case of cottonseed oil, it was assumed as 37 MJ/kg, the default value for vegetable oils in European legislation [7]; in the case of cottonseed meal, it was calculated from the properties of each component - lean meal, lintered hulls, oil (leaked). The heating value of de-oiled cottonseed meal and cotton linters were found in a report issued by the EU Joint Research Center (JRC), with values of 19.7 and 16.0 $^{MJ}/_{kg_{dry}}$ respectively [17]; the LHV of cottonseed hulls was found in an online database, with a value of 16 MJ/kg dry (interestingly measured from Israeli cottonseed) [113]. The moisture content of oil extraction solid outputs was set at 11% based on common practice for seed meals, which sets this value in the range of 10-12% [18] [78] [108]. The meal, in fact, typically exits the desolventiser at 18-20% relative moisture, to be dried thereafter in order to adjust its moisture to commercial levels [101].

4.3.3 Grain supplements for replacement of cottonseed

The life cycle studied in the present work features an oil extraction unit process with two co-products: cottonseed oil and cottonseed meal. Whenever two or more co-products are present, and no further subdivision of the process definitions can be implemented to distinguish the contributions of each flow, it becomes necessary to either apply system expansion or to allocate the environmental burden proportionally to a set criterium. In model EUC, the latter approach is adopted, coherently with EU guidelines on the matter; in the IS model, on the other hand, a system expansion up to the level of meal consumption is used. Consequently, this section interests only the construction of the IS model.

In our context, system expansion is a convenient approach to co-products accounting, because it is possible to clearly identify the substituted product; in many contexts, in fact, more or less plausible assumptions may be made on avoided products, accepting some degree of uncertainty - e.g. when a seed meal displaces another animal feed, it is common to assume that the displacement will happen at the expense of soybean meal. In this research, a baseline case in which whole cottonseed is delivered to cattle farms exists: consequently, the LCA unambiguously avoids the consumption of such wholeseed in this specific context, while providing cottonseed meal. This situation is beneficial for the accuracy of the study.

When whole cottonseed undergoes oil extraction, some nutritional value is lost in the process, as oil is removed from the seed. This means that, besides the mere substitution of whole cottonseed with cottonseed meal, it will be necessary to find a way to account for this nutrient loss via supply of feed supplements. As a common practice, the equivalence between different meals in a product substitution framework is made on the base of protein content, which represents the most important nutritional principle behind the commercial value of such animal feeds [15]. In this research, coherently with past research, two principles are adopted: feed energy and protein content [108] [114] [115]. Since feeding cottonseed is largely restricted to cattle due to the already mentioned toxicity issues, it is possible to use energy and protein content indicators that also account for the digestibility of the feed for this animal in particular. In the case of energy, the chosen indicator is known as Net Energy for Maintenance (NEm): it is defined as the amount of feed energy intake that will result in no net loss or gain of energy from bodily tissues, and it is measured in $^{Mcal}/_{kg_{dry}}$; any feed has a characteristic NEm value, accompanied also by a value of Net Energy for Growth (NEg), i.e. net energy for tissue growth: together, these two constitute the basis of the *net energy system* for cattle diet formulation. It is important to underline that NEm, which represents its ability in meeting the NEm requirement of the animal, is always slightly higher than the NEg value of the same feed, since the metabolic efficiency is always higher for maintenance than for growth. This research is based on NEm, since maintenance is the first priority for the well-being of the animal.

In the case of protein, the indicator adopted for the substitution is the quantity of Available Proteins (AP), measured in $^{kg}/_{kg_{dry}}$. The method using available proteins is based upon the use of crude proteins, i.e. total proteins by weight, by also accounting for their digestibility in cattle. This value characterises each feed according to formula 4.3.

$$AP_i = CP_i * pd_i \tag{4.3}$$

AP_i	$^{kg}/_{kg_{dry}}$	Available protein content of feed i
CP_i	$^{kg}/_{kg_{dry}}$	Crude protein content of feed i
d_i		Protein digestibility of feed i for cattle

The underlying assumption of this binary approach is that, besides these two macronutrients, the considered feeds are highly substitutable, i.e. there will be no appreciable impact on cattle nor on its milk yield after replacement. This is supported by the fact that oils and fats are not an essential component of cattle diet, differently from dietary fibres, energy and proteins. Oils can be totally excluded from the diet and, when used, it is recommended that they should not to surpass 2-3% of the total energy intake. Their use can even be counterproductive when feeding milking cows [116], although they may be intended as a feed supplement to support energy intake of early lactating cows [117]. Finally, note that, while crude proteins would be equal between wholeseed and its constituents if taken one by one, protein digestibility breaks this equivalence, meaning that the totals do not exactly coincide; in the case of cottonseed, digestibility of its single components is slightly lower than digestibility of wholeseed for cattle.

After the selection of these parameters, it was necessary to evaluate the nutrient content in cottonseed meal: this was done proportionally to the nutrient content and the dry mass share of each of its constituents, i.e. lean meal, lintered hulls and oil leaked during extraction. The needed nutritional data for cattle were taken from the literature [34]. Such nutritional values are modelled in function of leaked oil, i.e. of oil yield. The model considers the quantities of each meal constituent - lean meal, hulls, linters, leaked oil - coming out of the oil extraction unit and computes its nutritional properties proportionally to each constituent. Therefore, a more inefficient extraction unit will lead to a meal of higher energy value in the model.

The next step was to determine the feeds that would accompany cottonseed meal in order to supplement the missing nutrients; this was done based on the most recent import data in matter of grains in Israel, and focusing on cereals, which have proved to be the most suitable energy supplement feed. Israel, being a small and densely populated country, covers most of its food and feed demand with imports. Data on grain and oilseed imports for 2019 show that 73% of such imports were destined to the animal industry [106] and, what is more interesting, maize dominated in the feed industry, with 62% import share; feed wheat followed with 22%, and barley with 13%. Although the exact shares are subject to change yearly due to seasonal variations in market prices, it was assumed that such a hierarchy in import quantities would hold: the present study thus considers maize, wheat and barley grains as main feed supplements. Their relevant properties are reported in table 4.5.

Note that nutrient tables do not feature any dry matter composition of the seeds for

Table 4.5: Nutritional properties of each animal feed considered in the current research [34, p. 134-142].

Feed	**NEm** $[{}^{Mcal}/_{kg_{dry}}]$	**CP** $[{}^{kg}/_{kg_{dry}}]$	**pd** [%]	**AP** $[{}^{kg}/_{kg_{dry}}]$
Whole cottonseed	2.24	0.244	83	0.178
Cottonseed lean meal	1.79	0.461	57	0.263
Cottonseed hulls (lintered)	0.68	0.042	50	0.021
Cottonseed oil	4.75	0	0	0
Maize grains	2.24	0.098	45	0.0441
Wheat grains	2.18	0.142	77	0.109
Barley grains	2.06	0.281	73	0.096

which properties are reported; this, together with the fact that net energy of the seed cannot be deducted from the summation of its constituents, implies some unavoidable uncertainty in the feed supplement estimation. The table shows how maize, compared to the other grains, delivers slightly more energy, but at the cost of a much lower available protein content. Barley and wheat, on the other hand, show very similar macronutrient properties.

Regarding the origin of such feed imports in Israel, Ukraine stands at 44% market share; the EU and the USA follow with a 18% and 17% share respectively [106]. The issue of modelling supply from Ukraine, which would be ideal if looking at the import shares, is the total lack of reliable LCI data for crop cultivation in this country, a problem that has been cited also by the EU itself, as main importer of Ukrainian agricultural goods [118]; a single country-specific source was found, an online database from FAO [119], but the scarce precision of the data did not allow for its adoption.

Under these premises, it was decided to develop a base *feed supplement scheme* constituted by a mix of maize and wheat from the US. Such baseline features maize since it would reasonably be the marginal grain supplied to Israel, given its large margin over other grains in historical imports; since the EU is by far a net importer of maize, the US is the logical choice as geographical origin.

It is necessary to establish a methodology to assess the quantities of each of these two grain supplements, which have to replace missing nutrients in our analysis of the IS model. The goal of feed supplements is to cover the missing nutrients caused by the extraction of oil from the meal. The benchmark nutrient supply consists solely of wholeseed, and the alternative scenarios all feature cottonseed meal in some proportion to wholeseed, and one or more additional feeds. In our baseline feed supplement scheme, two feed grains are considered, namely maize and wheat. Here, the change in feeds caused by a shift from whole cottonseed to its meal was estimated through a simple set of two equations, as shown in 4.4a and 4.4b; here, $Feed_i$ represents the i-th feed dry mass.

$$NEm = \sum_{i}^{2}(Feed_{i,dry} * NEm_{i,dry}) \tag{4.4a}$$

$$AP = \sum_{i}^{2}(Feed_{i}dry * AP_i) \tag{4.4b}$$

The amounts of these two feeds resulting from the resolution of equations 4.4a and 4.4b for maize and wheat are shown in table 4.6, which reports such results in the baseline parametrisation of the model. The presented amounts of cottonseed meal, maize and wheat together supply the same amount of nutrients as 1 kg of wholeseed; in total, the resulting feed scheme features roughly 50 g of grains dry matter per kg of displaced cottonseed.

Table 4.6: Feed supplements and substitutions in use as baseline in the present study, per kg of whole cottonseed displaced.

Feed	**Quantity** $[kg_{dry}]$
Whole cottonseed	-1
Cottonseed meal	0.83
Maize grains (US)	0.10
Wheat grains (US)	0.41

In addition to this Baseline (BL) feed supplement scheme, two other alternatives have been developed: one investigates the LCA with barley from Denmark as feed supplement (scheme BDK); the other investigates the supply of feed wheat from the EU (scheme WEU). In the following paragraphs, we will discuss how the selection of agricultural sources has been carried out. In both these cases, the two-equation system is replaced with a single constraint tied to energy content. By testing the model over its range of oil yield and moisture contents, this approach has been proved to be solid for barley and wheat, whose nutrient content matches the needs caused by the lack of oil with proteins always in slight excess, from +1 to +5% with respect to the total protein requirement. [4] This compatibility is lost with maize, whose relatively lower protein content leads to larger excesses of grain and to changes in the limiting factor between NEm and AP. For this reason, feed supplement schemes including only maize were not modelled.

After selecting feed grains and computing their needs, it is fundamental to assess their environmental performance in terms of grain GWI. For this purpose, life-cycle data from different studies were collected and compared; this approach was considered necessary, as this research could not rely on a single, reliable source for feed grain LCIs. As already

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[4] This applies to all scenarios in the range of oil extraction yield of 90 to 99%, which is the relevant one for these schemes and solvent extraction, while for yields typical of simple press this would not be valid anymore.

mentioned, it was not possible to gather reliable data on Ukrainian grain production; thus, the inventory review focused on LCIs from the two locations following Ukraine by importance in Israeli imports of feeds, namely the US and the EU. The available LCA database, i.e. GaBi academic version - did not include any satisfactory LCI of grains. It was thus decided to rely on available literature: in this respect, many sources have been discarded because of missing information, in particular relating to the moisture content of the grain, or to the system boundaries definition.

After the review, eight studies were selected, the main features of which are shown in table 4.7. Three inventories deal with maize cultivation, three with barley and two with wheat. Before comparison, each source was adjusted on the functional unit of 1 kg dry matter of cereal.

Table 4.7: Main features of the data sources considered prior to developing the feed supplement schemes utilised in this research. M, B, W stand for maize, barley, and wheat, respectively.

Source	**Grain**	**Product system**	**Location (a)**	**Data gathering time**	**Data quality**	
NREL	M	cradle-to-farm-gate	US(Great Plains)	2005-2009	Producer survey and literature	[120]
NREL	M	cradle-to-farm-gate	US(Midwest)	2005-2009	Producer surveys and literature	[120]
(Kim,2009)	M	cradle-to-harvest	US(Corn Belt)	2000-2003	County and State-level databases	[121]
JRC	B	cradle-to-farm gate	EU	2012-2016	Consultancy and literature	[18]
DCA	B	cradle-to-harvest	DK	2016	Field measurements and literature	[122]
(Spatari,2020)	B	cradle-to-harvest	US	2014	Producer survey and literature	[123]
JRC	W	cradle-to-farm-gate	EU	2012-2016	Consultancy and literature	[18]
NREL	W	cradle-to-regional-storage	US	2010	Producer survey and literature data	[125]

(a) The Great Plains region is here intended as Texas, Oklahoma, Nebraska, Kansas, North Dakota, South Dakota, Montana, Idaho, Colorado, Wyoming, Utah, New Mexico, Arizona, and Nevada. Midwest is intended as Minnesota, Wisconsin, Michigan, Ohio, Indiana, Illinois, Missouri, and Iowa; the Corn Belt is made up of Illinois, Indiana, Iowa, Kansas, Kentucky, Michigan, Minnesota, Missouri, Nebraska, North and South Dakota, Ohio, and Wisconsin, thus being partially represented in both previous inventories. This will have some relevance when comparing the different datasets.

The sources of which in table 4.7 differ both in terms of system boundaries and of cut-off criteria. Prior to comparison, system boundaries were brought to a common ground via disaggregated data on after-harvest practices, which were available for the EU situation; this way, by also accounting for the different moisture contents, all inventories were related to the same functional unit (1 kg dry matter at the farm gate). In the case of wheat from the NREL database, values were brought back to farm gate for the comparison by accounting for the transport process by truck that all other American grains were supposed to undergo in the model, i.e. 150km by Euro4 20-26t truck; more about transportation processes can be found in section 4.3.5.

Some inputs are not accounted for, as their impact is deemed negligible with respect to the research goal and scope, and they are not always mentioned in inventories: these include pesticides, herbicides and fungicides, which are applied in tiny amounts compared to fertilisers - up to four orders of magnitude lower application rate than fertilisers [18] [125] [126]. While they may be cause of appreciable effects under impact categories linked, for example, with soil poisoning, they are assessed to have nearly no impact on GWI, and for this reason are often excluded from agricultural inventories [111] [127] [128]. Also seedling material was excluded from the inventories, as its impact is also irrelevant given our goal and scope.

In order to get a sense of the variability of agricultural inputs in feed grains, which will influence the GWI of feed supplements and thus the GHG intensity of cottonseed-oil RD under the ISO framework, inventories were compared in their most important inputs; this was carried out by group: maize was analysed separately, whereas wheat and barley were analysed together. Figure 4.7 shows the sources dealing with maize cultivation.

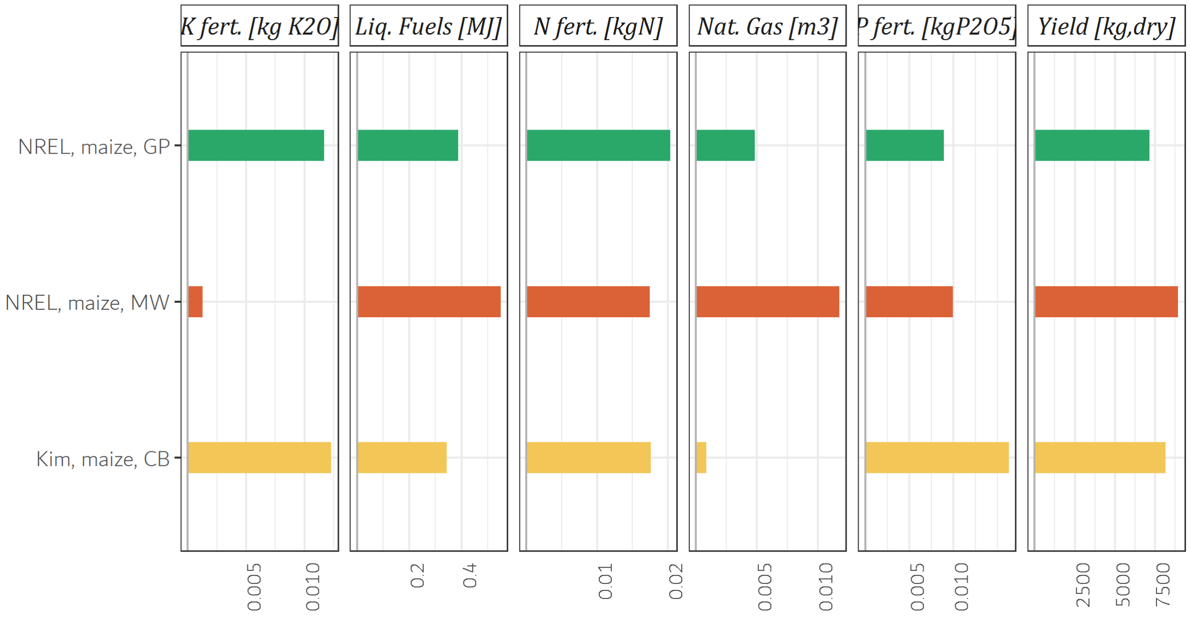

Figure 4.7: Comparison of the main agricultural inputs and yield among the considered maize product systems. All inputs are per kg of dry grain (GP=Great Plains; CB=Corn Belt).

In this figure, it is possible to notice a general agreement in fuels and nitrogen fertilisers inputs, which are by far the most impacting inputs in terms of GHG emissions. At the same time, appreciable relative differences can be found in K fertilisers input from Corn Belt NREL data and in natural gas inputs from Great Plains NREL data. The first discrepancy may be explained by the different K and P requirements that different soil types imply, whereas the latter may be partly explained by the difference in climate between the warmer Great Plains regions and the colder Midwest, which brings to different average drying requirements.

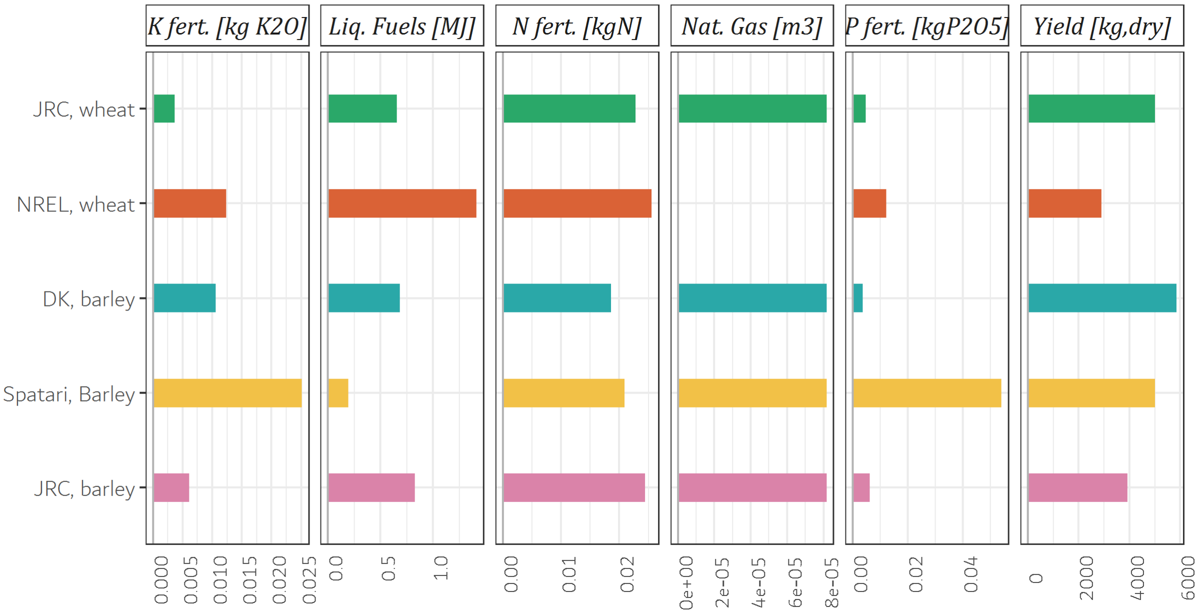

Figure 4.8: Comparison of the main agricultural inputs and yield among the considered wheat product systems. All inputs are per kg of dry grain.

Figure 4.8 shows how wheat and barley datasets are characterised by more variability. Nitrogen inputs are very close, whereas fuel and gas inputs highlights the difference between NREL wheat and all the other sources; the lack in natural gas use and the contextual higher reliance on liquid fuels may indicate a difference in the drying fuel. With regards to P and K fertilisers, barley from (Spatari,2020) features much higher consumptions, whereas the other datasets are more in line; this study also seems well below average in liquid fuels consumption. Finally, with regards to grain yield, European datasets all surpass 4 t/ha; American datasets, on the other hand, feature lower yields, which do not reach 3 t/ha in case of NREL wheat. This fact explains why NREL wheat tends to have higher inputs per kg of grain than most other sources.

Since US maize is primarily cultivated in the Corn Belt, it was decided to rely on data from this area, setting aside NREL the dataset from the Great Plains. Thereafter, it was decided to rely on the most conservative source of the two. The modelling of each emission source is discussed further on in this section: in this paragraph, results per unit mass of grain are briefly discussed just to justify the adoption of a certain dataset. System boundaries from (Kim,2009) were extended to account for on-farm drying and storage, using European data on the matter [18]; additionally, field emissions were accounted following the methodology explained later in this chapter. The resulting comparison shown that the GWI amounts to 284 and 339 ${}^{g_{CO2eq}}/_{kg_{dry}}$ of grain for (NREL) and (Kim,2009) respectively, so the latter source was selected.

US maize is represented by a relatively large number of studies, which made a comparison possible. By considering 12 additional studies, it was found that the average GWI amounts to 394 g_{CO2eq} per kg of dry grain, with a standard deviation of about 137 g_{CO2eq} [128]. [5] Our baseline case is thus circa 14% more efficient than the average US maize as in these studies; in any case, this difference is not going to play any important role in the results, since wheat prevails over maize, under all modelled circumstances, by a factor of around 4 to 5.

For US wheat, only one source is available, and there is no possibility of direct comparison with other American studies; other sources on US wheat, in fact, commonly lacked a transparent LCI, or did not clearly report any moisture content [129] [130].

As already mentioned, two additional feed supplement schemes were developed, both featuring Europe as source of grains. One relies on imports of Danish barley - scheme Danish Barley Scenario (BDK) - and one on European wheat - scheme EU Wheat Scenario (WEU); for this latter scheme, the dataset used is the one in (JRC,2017). This dataset features wheat targeted specifically at ethanol production, i.e. low-grade wheat and not durum or high-quality wheat of higher environmental impact. It is thus considered suitable for reflecting feed-grade wheat as well.

In the next paragraphs, the issue of agrochemicals modelling will be discussed. In this respect, fertilisers constitute the primary input in most agricultural systems. The three most important fertilisers have been included in the LCI: nitrogen (N), phosphorous (P) and potassium (K); it was deemed as necessary to improve the data quality on fertilisers as compared to what available in our database, following the approach outlined in the following paragraphs.

For N fertilisers life-cycle emissions, a first option would be relying on GaBi database information. However, this data is not specific to nitrogen content, and no information is given on the exact composition; furthermore, data are from 2001, too old to be reliable, given the progress in emission control and energy efficiency since then. In fact, while production techniques are roughly the same all across the globe, an appreciable difference in emission intensity can exist depending on: (I) the fossil fuel used as raw material for fertiliser synthesis; (II) the energy efficiency of the process; and (III) the efficiency of tail gas removal of the plant. For what concerns point (I), for instance, Chinese urea is usually synthesised via coal gasification, resulting in a footprint that is nearly twice the footprint of European, natural-gas-sourced urea. Concerning point (II), progress in energy efficiency, particularly of ammonia production - the first step in the production of most other N fertilisers - has led to consistent emissions savings in Europe [131]; but the most critical

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[5] Additionally to these 12 studies, literature offered also 3 studies reporting a negative GWI of maize. Negative GWIs are hardly explained without accounting for some degree of carbon accumulation in agricultural fields, which is not agreed upon in most sources. For this reason, and to keep the analysis as conservative as possible, these 3 sources were omitted to compute the GWI statistical distribution parameters.

factor that has to be considered when differentiating between European fertilisers and the rest of the world is the considerable progress in containing N_2O emissions at the plant level in nitric acid synthesis - with nitric acid being a key element in the chain leading to Ammonium Nitrate (AN), the most important fertiliser in Europe. The treatment of tail gas via heterogeneous catalysis allows for cutting in half the CO_{2eq} production emissions for nitric acid, thus reflecting on the emissions from its related fertilisers like AN [30].

In order to account for the aforementioned factors, nitrogen fertilisers were modelled separately for the EU and the US. It was not possible, in any case, to obtain crop-specific information with regards to the N-fertilisers in use, although this is supposed to have a secondary importance with respect to regional variations in fertiliser availability [131].

In the estimates for the EU, it was possible to distinguish between locally-sourced fertilisers and imports. Emission factors were sourced from published data by the International Fertilisers Society and by Yara Intl., the main EU producer [30] [131] and the worldwide leader in AN synthesis, distinguishing between local (EU) production and imports (from Russia, Africa and the Middle East); these data describe three relevant sources of nitrogen: urea, AN, Urea Ammonium Nitrate (UAN); the production of plain ammonia was instead described after European data, as contained in the GaBi database. These three represent together the totality of nitrogen applied to fields in the EU, according to JRC research commissioned by the EC [18]. By using the consumption shares as in the JRC report, while adopting more recent emission factors as in reports on fertilisers production emissions, the comparison with the results of the JRC differ by only 4%, with our estimate being lower [18].

All the relevant information concerning the emission factors (EF) are reported in table 4.8; the method does not allow to disaggregate emissions into the constituents - energy and fuel use, GHG leakages, etc. - but this level of aggregation, given the scope of the study, is deemed as acceptable.

In the case of the US, fertilisers supply is more fragmented. By relying on a USDA report documenting fertiliser use in the US from 1960 to 2015, it was possible to appreciate how, in the US, it is common to directly utilise anhydrous ammonia; in contrast with the EU, solid AN is not common at all, with more than 47% of 2012 consumption from UAN. These three fertilisers, together with urea, cover about 88% of the national demand [32]; the other sources of nitrogen - calcium ammonium nitrate, aqueous ammonia, nitrogen, ammonium sulphate etc. - were thus neglected. A second data source from the same agency allowed to trace the origin of imports [31]; the higher import dependence of the US fertilisers demand translates into a more diverse set of export markets, the main ones being Trinidad and Tobago, Canada, and Russia, but also with an appreciable share of imports from the Persian Gulf countries. Since consumption data on these substances are strictly related to final use as fertilisers, whereas available trade data entail all volumes regardless of the final use, it was not possible to determine the exact share of each imported fertiliser on the total amount of fertilisers used; it was thus deemed as sufficiently accurate to rely on declarations from the USDA, setting imported share of N fertilisers in final US consumption at 50% [132]. Of this 50%, a share is occupied by each fertiliser proportionally to its import volume. It thus results that, of total imports, nearly 55%

is anhydrous ammonia, and about one third is urea; the remaining 12% is covered by imports of UAN and AN.

The estimation of emission factor per unit nitrogen at the factory gate for US fertilisers proceeded by isolating the four most relevant fertilisers, and by considering all the countries making up 95% of US imports; then, importers were grouped by region aggregate, which is useful to relate them to a specific emission factor according to (ifs,2018) [30]. The source does not feature ammonia production emissions, which are caused mainly natural gas consumption, electricity consumption for the air separation unit and natural gas leakage; these were thus estimated based on available aggregated data for production in the EU, sourced from the GaBi database; this introduces a discrepancy, mainly rooted in the difference in electricity supply, which was not possible to disaggregate, but it is deemed as acceptable. As a counter-proof, this value was compared with estimates for the US from Fertilisers Europe, which, under the same conditions, result in just 4% more ammonia production emissions; the most conservative and documented dataset, i.e. the one sourced from GaBi, was adopted [133]. [6]

Table 4.8 contains the main information used to compute the average emission factors; the complete set of values used for the calculation of the US average fertiliser production emission factor is reported in annex.

As table 4.8 shows, the average EU fertiliser emits about 25% less per unit nitrogen than its US peer. Additionally to the impact at production plant gate, the impact related to transportation to the country of consumption was included in the analysis: for simplicity, only the main transport mode was accounted for - e.g. container ship from Trinidad and Tobago to the US or train from Russia to the EU; coherently with reports on the subject, such impact is minimal compared to the production emissions [134].

Fertilisers other than nitrogen are less critical to analyse. Phosphorous fertilisers are included in the GaBi database through one dataset, but it was deemed as incomplete: in fact, it models acidification emissions and eutrophication emissions, but it does not include any GHG emission. Furthermore, potassium fertilisers are not included at all in the available database: these two were thus modelled after a literature review.

In the case of P fertilisers, two data sources which do not account for capital goods were found: an older one, from the early 90's Western Europe, stating 1.19 kg_{CO2eq}/kg_{P2O5} [108], and a newer one, developed by Fertilisers Europe in 2015, stating 0.542 kg_{CO2eq}/kg at plant gate [18]: this last value is used in the current analysis, regardless of the country of production; note that in both the EU and the US, local production volumes allow not to rely on imports.

The same data source as above also provides a reference value for the European K fertiliser mix, at 0.417 kg_{CO2eq}/kg_{K2O}. In the US, K fertilisers are imported almost in their entirety from Canada [31]. Lime production data were taken from the GaBi database,

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[6] Ammonia emission factors account also for the emissions of the CO_2 captured during synthesis, which is invariably emitted soon after field application. Sometimes, these emissions are accounted to the agricultural stage instead.

Table 4.8: Composition and emission factor of the average N fertiliser used in the US and in the EU, as calculated in the present resource (system boundary at production plant gate).

Fertiliser	**Origin**	**Share** [a]	**EF** $\left[{}^{kg_{CO2eq}}/_{kg_N}\right]$
- US -			
Ammonia	Various	24.0%	5.18
Urea	Various	25.9%	3.74
UAN	Various	47.3%	5.20
AN	Various	2.8%	5.71
Average US fertiliser			**4.83**
- EU -			
Urea	EU	9%	3.50
Urea	Russia	11%	4.00
Urea	Africa/Middle East	16%	3.49[b]
AN	EU	59%	3.32
AN	Russia	5%	7.11
Average EU fertiliser			**3.63**

a) In percent points of total applied N.

b) Calculated as average between the factors of the two regions.

sourced from a German facility in 2019. The process is very similar regardless of the location, and most emissions are related to mining.

Together with grains, the agricultural systems considered deliver also secondary products such as stalks and stover; these field residues were invariably considered as by-product, i.e. all the environmental burden up to harvest was solely assigned to the grain. In most cases, the LCI for wheat ends after harvest: it was thus necessary to include additional unit processes of drying, storage and transport to a regional storehouse. Assuming that drying technology is similar between North America and the EU, JRC database information for drying and storage of wheat for ethanol were adopted [18]; these are reported in annex.

All the assumptions regarding transport are illustrated in the dedicated section 4.3.5; the same applies for the modelling of electricity supply, available in section 4.3.6.

All the datasets of which at table 4.7 feature field emissions for nitrous oxide N_2O, all estimated using different techniques in a more or less transparent way; while keeping track of the emission estimates for each dataset, the baseline emission factors utilised in this research have been uniformed by following IPCC tier 1 methodology; this ensured consistency between different feed supply scenarios, while adopting a conservative approach. A tier 2 method has been applied as alternative when geographical and soil information were sufficiently accurate, as shown in the results presented in section 5.2.5. In the following paragraphs, these two approaches are explained in detail.

The estimation of field emissions to air comprises a variety of mechanisms with different source and intensity. In the present research, the emissions considered are:

- N_2O emissions, both direct and indirect.
- CO_2 emissions from lime application.
- CO_2 emissions from urea decomposition.

Note that CO_2 emissions from urea decomposition when used as fertiliser, linked to carbon dioxide that is freed from soil after application, are not accounted in the agricultural phase, as the emission of CO_2 is allocated completely at the production site.

The most relevant GHG emitted by agricultural systems is nitrous oxide, owing to the potent effect that this gas has on radiative forcing - its GWI at 100 years time horizon makes it 298 times more impacting than CO_2 on weight base.

There are three pathways, one direct and two indirect, leading to field N_2O emissions. The direct pathway includes N_2O formation caused by the increase in available nitrogen in agricultural soils, which spurs nitrification and denitrification rates. The indirect pathways consist of: (I) the emissions following volatilisation of nitrogen in form of ammonia and NO_x, and their subsequent deposition, with production of nitrate (NO_3^-) and ammonium (NH_4^+) ions; and (II) emissions from leaching and runoff, caused by overland water flow - induced for instance by heavy rainfalls - or by flow through soil micropores and pipe drains; these phenomena lead to nitrogen - mainly in the form of (NO_3^- - bypassing the field, and partially forming nitrous oxide from the site in which the land drainage water eventually flows.

The total N_2O field emissions per unit area are calculated by summation of the contribution of direct and indirect emissions, as in equation 4.5.

$$E_{N2O} = E_{N2O,dir} + E_{N2O,indir} \tag{4.5}$$

As already mentioned, the framework for the estimation of these emissions is the one presented in 2006 IPCC guidelines [135]. Said methodology distinguishes three tiers of methods to be used, depending on the quantity and quality of available data to support the estimation. Equations from the IPCC report have been slightly adjusted in order to adapt them to our case, and in order to describe both the tier 1 and the tier 2 approach with a single, uniform explanation when possible.

The general formula used for the estimation of direct N_2O emissions, valid for both tier 1 and tier 2 approaches, is shown as equation 4.6.

$$E_{N2O,dir} = E_{dir,applN} + F_{CR} * EF_1 * {}^{44}\!/_{28} \tag{4.6}$$

E_{N2O}	kgN_2O ha^{-1}a^{-1}	total annual N_2O emissions
$E_{N2O,dir}$	kgN_2O ha^{-1}a^{-1}	annual direct N_2O emissions
$E_{N2O,indir}$	kgN_2O ha^{-1}a^{-1}	annual indirect N_2O emissions
$E_{dir,applN}$	kgN_2O ha^{-1}a^{-1}	annual direct emissions due to nitrogen application
F_{CR}	kgN ha^{-1}a^{-1}	annual amount of N in crop residues
EF_1	0.01 kgN_2O-N kg N^{-1}	tier 1 emission factor for direct N_2O emissions
${}^{44}\!/_{28}$	kgN_2O kgN_2O-N^{-1}	conversion factor

Direct emissions are disaggregated into emissions from nitrogen in fertilisers, and emissions linked to nitrogen stored in crop residues left on field. Emissions from residues are computed separately from the rest of the emissions, using a tier 1 approach; the tier 2 approach applies to all other N sources, i.e. synthetic fertilisers and organic fertilisers. In principle, this calculation must also account for the additional losses induced by farming on organic soils, but, since this research features exclusively cultivation on mineral soils, it can be omitted.

Equations 4.7a and 4.7b show how $E_{dir,applN}$ was computed according to each approach, i.e. for tier 1 and tier 2 respectively.

$$E_{dir,applN} = F_{SN} * EF_1 * {}^{44}\!/_{28} \tag{4.7a}$$

$$\begin{aligned} E_{dir,applN} &= exp(-1.516 + 0.0038 * (F_{SN} + F_{ON}) + ev_{soc} \\ &+ ev_{pH} + ev_{tex} + ev_{clim} + ev_{veg} + ev_{expl}) - exp(-1.516 \\ &+ ev_{soc} + ev_{pH} + ev_{tex} + ev_{clim} + ev_{veg} + ev_{expl}) \end{aligned} \tag{4.7b}$$

F_{SN}	annual amount of synthetic fertilisers applied, in $[kg_N ha^{-1} a^{-1}]$
F_{ON}	annual amount of organic fertilisers applied, in $[kg_N ha^{-1} a^{-1}]$
ev_{soc}	soil organic C content parameter
ev_{pH}	soil pH parameter
ev_{tex}	soil texture parameter
ev_{clim}	climate zone parameter
ev_{veg}	vegetation class parameter
ev_{expl}	length of experiment parameter

The tier 1 method, although presented in a different parametrisation to be consistent with the other method utilised, is identical to the one presented in (IPCC,2006) [135]. In its essence, it makes use of a default emission factor of 0.01, implying that 1% of applied nitrogen is released as N_2O after application; this values is accompanied by a rather large uncertainty, which will be discussed in section 5.2.

The tier 2 method is based on a statistical model developed by Stehfest and Bouwman (2006), and it interests just direct N_2O emissions from fertilisers; it is the same model used in the calculation of GHG emission factors in European regulations. It bases its estimates on 1008 field measurements under varying environmental conditions, grouping results by crop type (cereals, grass, legume, other, wetland rice) [105]. It was possible to adopt this approach only in the feed supply scheme with Danish barley, BDK, since the study located barley cultivation in Western Denmark, while providing sufficiently accurate soil and climate data. At the base of this approach are six parameters, called "effect values", which characterise equation 4.7b. By means of these effect values, climate and soil parameters translate into a certain nitrous oxide emission intensity. For each parameter, the model features a series of thresholds that allow to assign a value to each effect value; in annex, it is possible to consult the detailed values and thresholds for each effect value. Table 4.9, instead, shows the particular values assumed by said parameters in the BDK feed supply scheme. These values were estimated by relying both on crop-specific and geography-specific information, both from the study itself and from the Global crop and site specific Nitrous Oxide emission Calculator (GNOC) database: this last source, which is available for online consultation, allows for a spatial resolution of about 10x10 km [136].

This method results in emissions at 1.41 $^{kg_{N2O}}/_{ha}$, about 18% lower than its tier 1 counterpart.

Table 4.9: Effect values for barley from Western Denmark, as adopted in the BDK feed supply scheme.

Effect value	**Value**
ev_{soc}	0.056
ev_{pH}	0
ev_{tex}	0
ev_{clim}	0.0226
ev_{veg}	0
ev_{expl}	1.991

All emissions other than direct N_2O from fertilisers have been invariably estimated using a tier 1 approach. Emissions caused by nitrogen in crop residues rely on the emission factor EF_1 applied to the amount of nitrogen in crop residues left on field F_{CR}. In order to estimate F_{CR}, the formula shown in equation 4.8 was adopted - as outlined in equation 11.7A from (IPCC,2006) [135].

$$\begin{split} F_{CR} = (1 - Frac_{burnt} * C_f) * AG_{DM} * N_{AG} * (1 - Frac_{remove}) \\ + (AG_{DM} + Yield_{dry}) * R_{BGbio} * N_{BG} \end{split} \tag{4.8}$$

$Frac_{burnt}$	/	Fraction of burned residue
C_f	/	Combustion factor
AG_{DM}	$t_{dry}ha^{-1}$	dry above-ground residues
N_{AG}	$kg_N kg_{dry}^{-1}$	N content of above-ground residues
$Frac_{remove}$	/	fraction of residue removed from field
BG_{DM}	$t_{dry}ha^{-1}$	dry below-ground residues
$Yield_{dry}$	$kgha^{-1}a^{-1}$	crop yield, dry weight
R_{BGbio}	/	ratio of below-ground to above-ground residues
N_{BG}	$kg_N kg_{dry}^{-1}$	N content of below-ground residues

The estimation of AG_{DM}, necessary in the calculation, is based on equation 4.9. It necessitates crop-specific parameters a and b - included in (IPCC,2006) - and dry matter yield $Yield_{dry}$. For cereals, the uncertainty on the crop-specific parameters used is very low, in the range of 3% (maize) to 8% (barley) [135].

$$AG_{DM} = \frac{Yield * DRY}{1000} * b + a \tag{4.9}$$

In the case of cereals, equation 4.8 mainly relies on crop-specific default factors of which in (IPCC,2006); such data were complemented by data on yield, sourced from the respective data sources, to determine $Yield_{dry}$.

The GNOC database provided instead the estimates for both $Frac_{burnt}$ and $Frac_{remove}$, based on geographical area and crop type. C_f is sourced from IPCC guidelines, assumed equal to 0.9 - the default value for wheat - for both wheat and barley. The complete set of values is reported in table 4.10.

Additionally to direct emissions, indirect emissions contribute appreciably to total field emissions of N_2O. Indirect emissions have been uniformly computed using the IPCC tier 1 method, which computes emissions based on the amount of N input from fertilisers and from N in crop residues. In principle, emissions from leaching should be included only for locations subject to occasional overcoming of soil water-holding capacity.

Leaching occurs in locations subject to occasional abundant precipitations and/or wherever irrigation is employed [135]. Since information on irrigation system and exact location was not always available, and in the logic of a conservative approach, this term was always included in the calculation. In particular, by relying on the GNOC database, it was observed that territories included in the study on maize used as source featured a mix of areas subject to rainfall-driven leaching and areas without leaching. The database also reported that Western Denmark - home of the study for Danish barley - is also subject to rainfall-driven leaching, while data on EU wheat cover the entire Union territory, and it was thus decided to follow a conservative approach and account for leaching as well. Data on US wheat are also a national average, and the same approach was thus applied.

In this case, all the values for emission factors and fractions have been taken from

Table 4.10: All the parameters used to estimate direct field emissions from crop residues.

Parameter	Unit	Maize	Barley	Wheat	Comment
$Frac_{burnt}$	/	0.03	0.03	0.03	Default value, from GNOC database [136]
C_f	/	0.8	0.9	0.9	Default values, crop-specific, for maize and wheat respectively [135].
AG_{DM}	$kg_{dry}ha^{-1}$	7990	6340	8090	Calculated based on dry matter yield and crop-specific parameters, as in equation 4.9. Here reported only for the three specific datasets of crops included in the scenarios.
N_{AG}	$kg_N kg_{dry}^{-1}$	0.007	0.006	0.006	Default values, crop-specific [135].
N_{BG}	$kg_N kg_{dry}^{-1}$	0.007	0.007	0.006	Default values, crop-specific [135].
$Frac_{remove}$	/	45.7%	20%	20%	Datum for maize extracted from the study, as average of the reported values of residue removal [121]. Data for maize and wheat are default, specific for Europe, taken from the GNOC database [136].
R_{BGbio}	/	22%	22%	24%	Default values, crop-specific [135].

(IPCC,2006). The approach is shown in equations 4.10.

$$E_{N2O,indir} = E_{vol} + E_{leach} \tag{4.10}$$

$$E_{vol} = (F_{SN} * Frac_{GASF} + F_{ON} * Frac_{GASM}) * EF_4 * {}^{44}/_{28} \tag{4.11}$$

$$E_{leach} = (F_{SN} + F_{ON} + F_{CR}) * Frac_{leach} * EF_5 * {}^{44}/_{28} \tag{4.12}$$

E_{vol}	$kg_{N2O}ha^{-1}a^{-1}$	indirect emissions from volatilisation and deposition
E_{leach}	$kg_{N2O}ha^{-1}a^{-1}$	indirect emissions from leaching and runoff
$Frac_{GASF}$	$0.10\ kg_{NH3-N+NOx-N}kg_N^{-1}$	volatilisation from synthetic fertilisers
$Frac_{GASM}$	$0.20\ kg_{NH3-N+NOx-N}kg_N^{-1}$	volatilisation from organic fertilisers
$Frac_{leach}$	$0.30\ kg_N kg_N^{-1}$	N losses by leaching/runoff in areas where it occurs
EF_4	$0.01\ kg_{N2O-N}kg_{NH3-N+NOx-N}^{-1}$	emission factor for volatilisation and deposition
EF_5	$0.0075\ kg_{N2O-N}kg_N^{-1}$	emission factor for leaching/runoff

The last agricultural input to be considered is lime. The use of lime in agriculture serves the function of neutralising the acids that form in the soil after application of N fertilisers, in order to prevent the soil pH from lowering. CO_2 emissions originate via the neutralisation reaction shown in equation 4.13.

$$2HNO_3 + CaCO_3 \rightleftharpoons Ca^{++} + 2(NO_3^-) + CO_2 + H_2O \tag{4.13}$$

These emissions are accounted in the fertilisers, as they are ultimately caused by fertiliser application, and as they would occur even by reaction of carbonates already present in the soil. The approach used for the calculation of such emissions is again the tier 1 approach, as the most conservative among all the methods: in fact, it considers emission factors equal to the total carbon content in lime; the procedure is shown in equation 4.14.

$$E_{lime} = M_{lime} * EF_{lime} * {}^{44}\!/_{12} \tag{4.14}$$

E_{lime}	$kg_{CO2}ha^{-1}a^{-1}$	total CO_2 emissions from liming
M_{lime}	$kg_{lime}ha^{-1}a^{-1}$	annual amount of lime applied to soils
EF_{lime}	$0.12\ kg_{C}kg_{lime}^{-1}$	emission factor for limestone
${}^{44}\!/_{12}$	$kg_{CO2}kg_{C}^{-1}$	conversion factor

Since it is difficult to find disaggregated data between limestone ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$), and their emission factors differ only by 0.01, all lime applied is considered to be limestone under all circumstances.

The last aspect of grain supplements to take into consideration when modelling their GHG impact is related to the ILUC effects of an increase in their demand. Owing to its rather complex nature, this topic has been addressed in a dedicated section, namely section 4.3.7.

4.3.4 Hydroconversion of cottonseed oil

As illustrated in section 3.2.3, the Ecofining unit featured in this study is made up of a stage of pre-treatment, an hydroconversion section - with one reactor for hydrogenation and oxygen removal and one for isomerisation - and a simple fractionation section. Additionally, the plant includes side-processes such as hydrogen recycling and amine scrubbing for CO_2 removal. In the following paragraphs, material and energy inputs will be treated separately, as different approaches have been adopted for these two instances.

With regards to utilities consumption, although it was not possible to find first-hand industrial data from an actual Ecofining unit, it was possible to rely on models of such plants. A model of an Ecofining unit was developed by the Argonne National Laboratory using Aspen Plus, which states heat and electricity needs at the highest level of aggregation, i.e. including pre-treatment [16]. Another study on Ecofining, which relies on the same source as well as on consultancy with an expert at UOP, assumes about the same electricity consumption, and a heat need about 4% higher to produce RD [137]; as the latter are slightly more conservative, it was chosen to use its values. Both sources have a limitation: they model HVO production without considering the isomerisation process. Notwithstanding, the second of these sources reports an additional consumption of +10 to +30% when producing jet fuel as main output instead of diesel fuel. Owing to the fact that jet fuel production implies a process of hydrocracking, similar but much more energy-demanding than hydroisomerisation, this information allows to assess additional consumption due to isomerisation certainly below this threshold. and this will be the threshold investigated in the parameter variation (i.e. +10% specific consumption). Per se, isomerisation of paraffins is characterised by a very mild exothermicity and a very low hydrogen consumption, which places its utility consumption well below the main hydroconversion reactor.

H_2 is supplied to the modelled Ecofining plant by means of a steam reforming unit. Natural gas is supplied as in the model of the Italian gas grid, and electricity is supplied as coming from the Italian electricity grid, in its average production mix. Process steam is supplied by a natural gas boiler at 90% efficiency. While hydrogen consumption is modelled starting from the chemical pathways inside the reactors, process heat and electricity consumption are estimated for the whole process, relying on literature data.

The material balance for the pre-treatment unit was based on industrial data for cottonseed, from a Brazilian study [2]. It is found that about 2.85% of incoming raw vegetable oil is lost, exiting as soapstock - phospholipids being the main contaminant in vegetable oils; the presence of free fatty acids is harmless, and even preferable, for the successive hydroconversion processes. Note that this value is consistent with the composition of 95 to 97% triglycerides indicated in literature on vegetable oil biorefining [10]. Inputs of chemicals - acids and bleaching earth - are excluded from the analysis, as their quantities are risible and their overall GHG impact is also minimal [109]; this is consistent also with other studies on hydroconversion of vegetable oils, and with many models for HVO LCA [16] [137] [138] [139].

Downstream of the pre-treatment unit, a first reactor has the purposes of breaking the oxygen bridge in the triglycerides, thus producing propane and fatty acids, and hydrogenating the double bonds in the fatty acids chains to produce n-paraffins; a second reactor serves the main purpose of enhancing the cold flow properties of the paraffinic mix, and can be tuned to produce high yields of gas oil or kerosene by adjusting the severity of the process and regulating cracking reactions. These require a more refined modelling than the pre-treatment unit, as outlined in the next paragraphs.

Input and output material flows, including hydrogen consumption, were modelled in a parametric way; the model makes use of the parameters (I) selectivity towards HDO S_{HDO} and final naphtha yield (by weight) Y_{naph} to provide its material balance. [7] This concept is synthetically visualised as black box in figure 4.9.

In order to link a selectivity to a product composition, two reactions of hydroconversion have been modelled, namely hydrodeoxygenation (HDO) and decarboxylation (DCO2), as shown in equation 3.3a and 3.3c while describing the chemistry of vegetable oil hydroconversion. There is no need to define the third reaction, i.e. decarbonylation (DCO), as the final product range can be related to two linearly independent reaction;

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[7] When referring to yields, the present text invariably implies a yield by weight, i.e. in terms of kg/kg_{feed}.

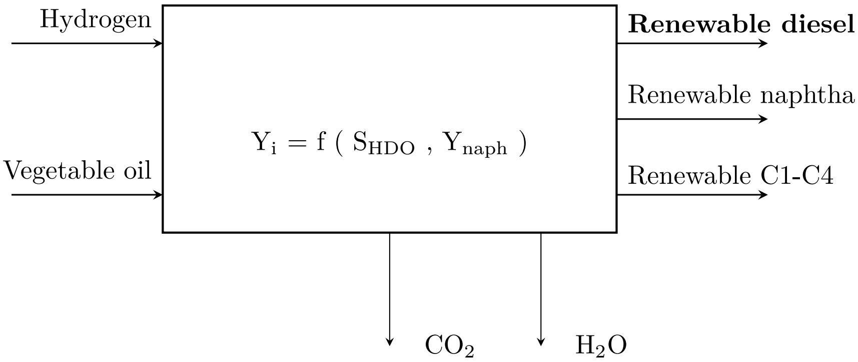

Figure 4.9: Schematic representation of relevant material flows and the functional relationship describing the yield of each product.

physically, what happens in the reactor is the conversion of CO to CO_2 via water gas shift, which is thermodynamically favoured [10].

Starting from the fatty acid composition of cottonseed oil of which at table 3.1, it was possible to determine the product yield of each n-alkane under complete conversion, for each reaction - HDO and DCO2 - separately. The hypothesis of complete conversion, which is applied also to the whole reactor model, matches the product yield in industrial practice [33]; note that the presence of FFAs in vegetable oil, normally in the range of 0.05% by weight, has been neglected, so that the oil can be considered as pure triglycerides.

Under such conditions, for each separate reaction of HDO and DCO2, it was possible to determine yields and mass fractions of the various n-paraffins, theoretical hydrogen consumption, and LHV of the paraffinic product. An interesting result is the theoretical hydrogen consumption (which depends solely on fatty acid composition): the mass fraction of H_2 was found to be 3.53% for pure HDO and 1.53% for DCO2; compared to soybean oil and palm oil, two common oil sources for biofuels other common feedstocks for HVO, cottonseed oil stands in the middle as far as hydrogen consumption is concerned. This can be seen for the HDO reaction in table 4.11. The full product compositions following HDO and DCO2 pathways can instead be consulted in the appendices.

Table 4.11: Comparison of HDO theoretical hydrogen consumption of cottonseed oil with the most common vegetable oil feedstocks.

Oil feed	**Stoichiometric H_2 weight fraction**
Palm oil	3.15%
Rapeseed oil	3.39%
Cottonseed oil	3.53%
Soybean oil	3.66%

Now that each reaction has been defined and tested for different oils, the choice of a selectivity S_{HDO}, together with the hypothesis of complete conversion, allows to determine product yields and hydrogen requirements of the first reactor, performing hydrogenation (note that once set S_{HDO}, it is immediate to find that $S_{DCO2} = 1 - S_{HDO}$). Here, the parametrisation of selectivity reflects two main instances, both discussed in section 3.2.2: (I) the role of catalyst choice, and (II) reaction conditions - mainly temperature.

The second parameter, Y_{naph}, is linked with the second reactor, and particularly to the severity of cracking reactions during isomerisation. In order to maximise diesel yield, cracking must be minimised, mainly by limiting the temperature and by an optimal catalyst choice; the trade-off in temperature is dictated by the kinetics of isomerisation, as higher temperatures also allow for a faster reaction and, therefore, lower residence times. Besides naphtha, cracking also generates an amount of lighter products, in the range C_1-C_4.

The reactor model requires a criterium to estimate the final mass fraction of C_1-C_4 based on the parameter of naphtha yield Y_{naph}. The estimation of such a correlation is made especially critical by the total lack of data regarding cottonseed-oil hydroconversion. As a consequence, the starting point for this analysis was the consideration of available data on product composition of Ecofining for palm oil and soybean oil, here reported in table 4.12.

Table 4.12: Product yield of palm and soybean oil through Ecofining, depending on the final diesel cloud point [10].

	Palm oil		**Soybean oil**	
Cloud point	0°C	-8°C	0°C	-8°C
CO_2	5.4	5.4	5.4	5.4
H_2O	8.3	8.3	8.3	8.3
C_1-C_4	5.9	6.9	6.6	7.0
Naphtha	2.9	9.5	7.7	9.6
Diesel	80.3	75.5	75.5	73

These data for palm oil and soybean oil were related to cottonseed oil based on the following considerations:

- *Longer n-paraffins*, i.e. with higher carbon-chain length, have a higher cloud point - with T_{cloud} correlated with carbon-chain length through a logarithmic trend (see figure 3.9), and this implies that *more isomerisation is necessary* once a desired final T_{cloud} is fixed [10].
- *Longer n-paraffins* exhibit a *higher reactivity to cracking* reactions, generally implying more abundance of light products after fixing the final T_{cloud} [140].

These two factors, which have a synergic effect on cracking occurrence, both suggest that carbon-chain length can be a good predictor of this phenomenon.

Furthermore, table 4.12 shows that the influence of initial paraffinic chain length (higher in soybean and lower in palm oil) is more evident when the desired T_{cloud} is higher, i.e. when the product does not undergo severe treatment: in this case, diesel yield is appreciably higher for shorter chain lengths.

These considerations on carbon-chain length allow to assume that treating cottonseed oil would require a level of isomerisation in-between palm and soybean oil, and would imply a degree of cracking in-between these two. The average carbon-chain length of cottonseed oil, in fact, is 17.46, roughly midway between the composition of palm and soybean oil.

The data in table 4.12 were thus interpolated for each of the two vegetable oils; a third point was calculated and added to each set prior to interpolation, representing c1-c4 yield (in the form of pure propane yield) at null naphtha yield $Y_{naph,0}$, i.e. representing the product composition if the isomerisation reactor were totally skipped. For each of these two sets of three points, a linear trend correlating naphtha yield Y_{naph} and C_1-C_4 yield was obtained, as shown in figure 4.10; for soybean, this results in a perfect fitting, whereas for palm oil the correlation is not as strong, but still solid for the purpose of this research.

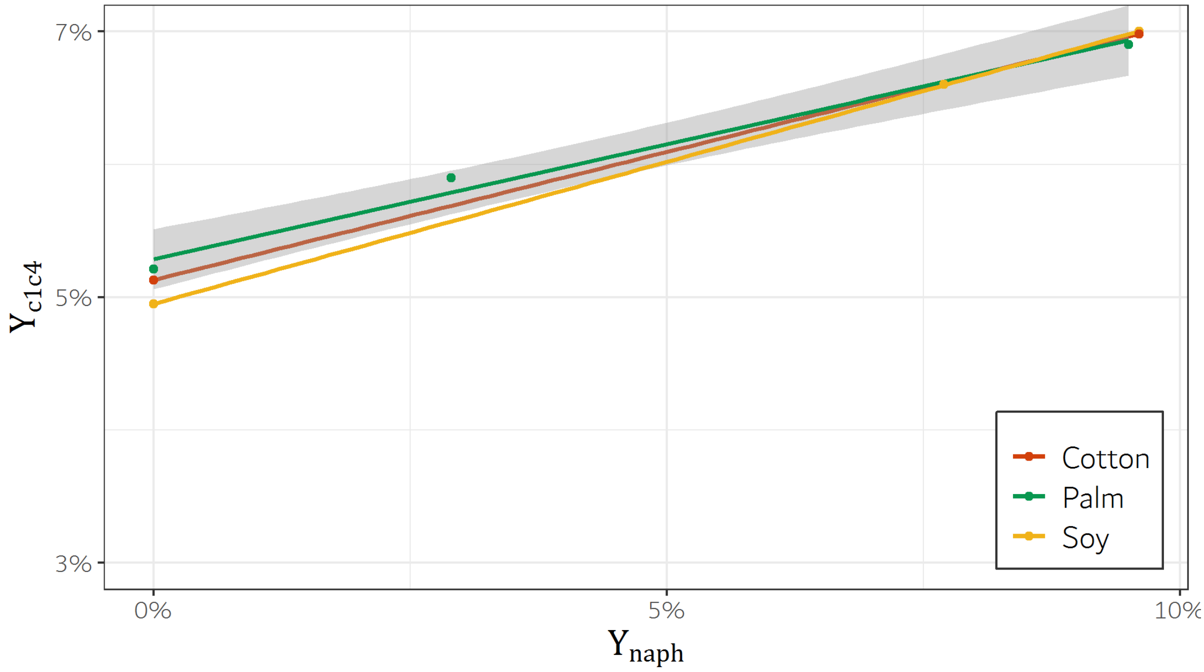

Figure 4.10: Correlation between naphtha yield and light products (C1-C4) for palm oil, soybean oil and cottonseed oil. The 95% confidence interval for palm oil is shown in grey.

The correlation for cottonseed was finally obtained by weighting the values of the $(\frac{Y_{C1-C4}}{Y_{naph}})$ ratio according to its carbon-chain length, as in equation 4.15. In order to then trace the trend for cotton as in figure 4.10, the information about propane yield at null naphtha formation $Y_{naph,0}$ was utilised together with the value obtained by the equation, effectively an angular coefficient.

$$c_{cott} = c_{palm} + (c_{soy} - c_{palm})\frac{l_{avg,cott} - l_{avg,palm}}{l_{avg,soy} - l_{avg,palm}} \tag{4.15}$$

Table 4.13: Average carbon-chain length and cracking products correlations between naphtha and C1-C4 produced in the isomerisation reactor.

Feedstock	l_{avg}	c $\left[{}^{kg_{c1c4,cracking}}/_{kg_{naph}}\right]$
Palm oil	17.06	0.1734
Soybean oil	17.45	0.2138
Cottonseed oil	17.86	0.1929

Thanks to this estimate, it is possible to link Y_{naph} to a value for Y_{C1-C4}: the value for c_{cott} suggests that light products are formed in a proportion of about 1 to 5 with respect to naphtha formation. This method cannot substitute the precision of direct measurements, but it is deemed as sufficiently accurate given the goal and scope of the research.

H_2 consumption in the second reactor $m_{H2,R2}$ is small compared to the consumption for hydrogenation; nonetheless, its input was considered in the analysis. After setting Y_{naph} and consequently also Y_{C1-C4}, a shift to extensive quantities allowed to calculate $m_{H2,R2}$, by considering the simple stoichiometry of hydrocracking; results show that this hydrogen consumption in the isomerisation reactor does not exceed some percentage points (1-4%) of $m_{H2,r1}$.

During the modelling of this section of the plant, a discrepancy between all hydroconversion plants as modelled in academic studies and industrial data and textbooks was detected, a discrepancy concerning product composition. The model developed so far shows that, if selectivity is tuned for: (I) an exclusive presence of HDO reaction, i.e. $S_{HDO} = 1$; and (II) a purely theoretic absent naphtha production, the diesel yield Y_{diesel} for a triglyceride feed stands at a value of 85.9%; with $Y_{naph} = 1\%$, Y_{diesel} drops to 84.8%.

Textbooks on Ecofining normally report a diesel output of 75 to 85 wt%, as in table 3.3, and these same sources report a naphtha yield of 1 to 8 wt% [10] [33]. The model seems thus to match the expected maximum, although this value is purely theoretical, hardly attainable in real reactors and under the usual fuel specification requirements. Published studies on HVO, on the other hand, were found to feature diesel yields around this value: an LCA based on the NExBTL process reports 83.9%, and a naphtha yield of just 2.1 wt% [141]; two studies dealing with Ecofining report RD yields at 85.1 and 84.2 wt% respectively [16] [137]; the first of these studies dealt with soybean oil, whereas the second one with a generic vegetable oil. Finally, in its work for the EC, the JRC reported a Y_{diesel} of 84.8% for a generic vegetable oil. All these values, which are very close to the ideal yield with minimal naphtha formation, imply a very low naphtha production and also very high selectivity towards HDO. However, as table 4.12 points out, reaching a final cloud point of 0°C with palm oil would yield about 3% naphtha, yielding about 82.5% paraffins under complete conversion; furthermore, admitting for some presence of

DCO2 reaction would further lower this result. All values reported above exceed this one, thus implying either a total absence or a very limited intensity of isomerisation. This aspect leads to an overestimation of yields, especially when dealing with oils of appreciable unsaturation like soybean oil. In this respect, most studies are not based on the actual triglyceride composition of the feedstock, which is important to determine the specific hydrogen consumption: this leads to further underestimation of this process in terms of material requirements and impacts.

Naphtha yield will be sensitive to the final target cloud point of the fuel, and with it the diesel yield: in practical terms, a reasonable naphtha yield to be expected lies on the higher end of the product yields as in table 3.3, i.e. 7 to 8%. Following consultation with an expert at the company operating both Italian HVO biorefineries, the industrial final naphtha yield is in the range of 7 to 13% [142]. Two pieces of information supported the determination of S_{HDO} and Y_{naph}: (I) the information on CO_2 and H_2O yields as from table T1 (which are all identical), where CO_2 can be considered a marker of DCO2 occurrence and H_2O of HDO; and (II) the fact, cited in the literature [10], that the reactor uses a mixed catalyst which is reactive to both HDO and DCO2. Based on this set of information, it was possible to tune the baseline case at $S_{HDO} = 65\%$ and $Y_{naph} = 8.5\%$. This value of selectivity yields CO_2 and H_2O at 5.4% and 8.2% respectively, very close to the industrial data for these chemical species of which in table 4.12. With these values, diesel yield stands at 74.4%, and hydrogen consumption stands at just above 3%. Note that such a value of naphtha yield reflects final diesel cloud points in the proximity of -5 to -8°C; this implies a diesel that is generally used in winter, when lower cloud points are required: for diesel produced in Northern Europe, for instance, a cloud point of -7°C in winter and +2°C in summer is reported in the literature [13].

In figure 4.11, it is possible to compare this setup with other setups, depending on the value of the two parameters. As already pointed out, different selectivities in the graph correspond to differences mainly in terms of catalyst and reaction temperature; differences in naphtha yield depend on catalyst and temperature as well, being linked to different occurrence of cracking and thus different target in cold flow properties.

Setup	$\mathbf{S_{HDO}}$	$\mathbf{Y_{naph}}$	$\mathbf{Y_{diesel}}$	$\mathbf{m_{H2}}$ $[kg/t_{feed}]$	$\mathbf{m_{H2,r2}/m_{H2,r1}}$
1	0%	1%	79.9%	15.7	0.81%
2	100%	1%	84.8%	36.7	0.35%
3	0%	13%	66.0%	17.2	10.56%
4	100%	13%	70.9%	38.2	4.50%
B	65%	8.5%	74.4%	30.3	3.68%

The table related to figure 4.11 reports also extensive hydrogen consumption and the ratio of consumption between the two reactors. A minimum of 15.7 kg_{H2}/t_{feed} is consumed when minimising naphtha yield and silencing the DCO2 reaction, whereas a maximum of 36.7 kg_{H2}/t_{feed} is consumed under full DCO2 and maximum naphtha formation.

In the reactor model, leakages of hydrogen or any other gas, e.g. from the amine

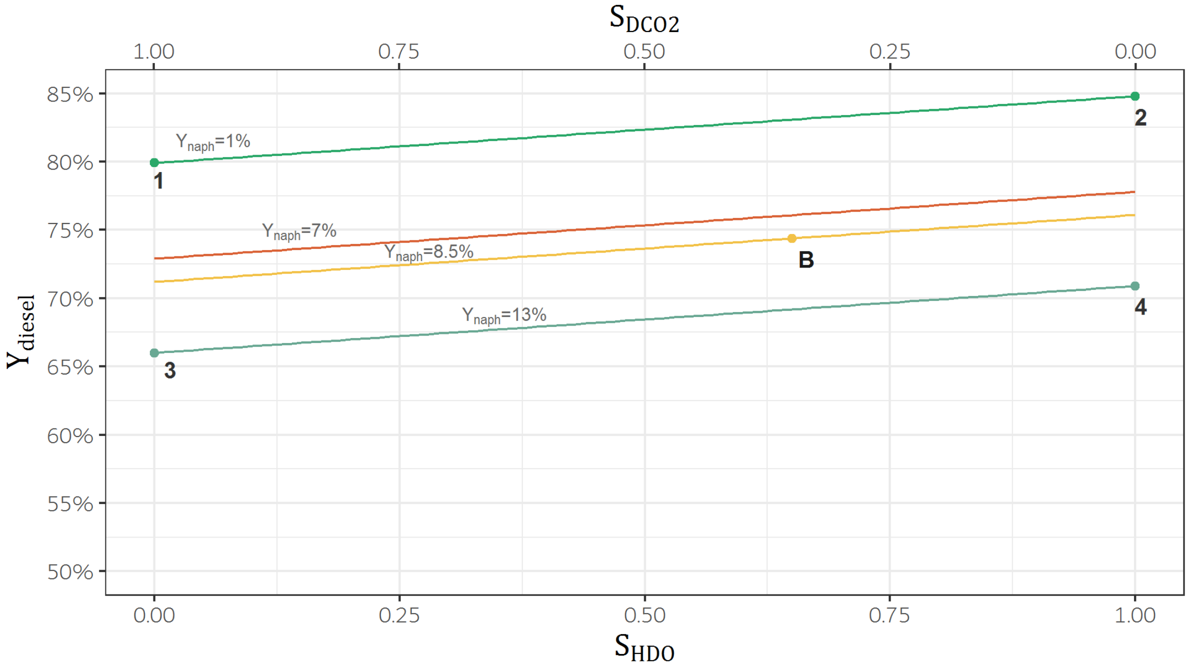

Figure 4.11: Diesel yield depending on HDO reaction selectivity, with varying final naphtha yield.

scrubber, are not evaluated and thus neglected, consistently with the goal and scope of the study.

The complete inventory data in the aforementioned conditions, i.e. as in point B in figure 4.11, are reported in table 4.14. These are the ones used as baseline for all considered scenarios, and they will be tested in section 5.2.

Table 4.14: Inventory data for the biorefining process step, per ton of feed.

Inputs		
Oil feed	kg	1000
Hydrogen	kg	30.17
Electricity	kWh	46.14
Heat (steam)	MJ	148.79
Outputs		
Diesel	kg	740.60
Naphtha	kg	87.56
C_1-C_4	kg	14.99
CO_2	kg	53.87
H_2O	kg	81.85

4.3.5 Transport processes

Transport processes exist all along the product system; land transport links the various steps leading to cottonseed oil and grains shipping, and naval transport covers the remaining distances. These processes were modelled with the support of database information on the various transportation modes. In GaBi, adaptable parameters in transportation datasets are: distance, utilisation ratio (also known as load factor), share of road categories, required sulphur content and share of biogenic C in fuel; the most impacting parameters for transportation emissions are distance and utilisation ratio.

The utilisation ratio represents how well the cargo capacity of a transport means is exploited: for instance, a truck loaded to its full load has a unitary utilisation ratio. In this research, this parameter accounts at once also for the return trip (round-trip utilisation ratio). For naval transport, which was considered to be carried out by means of bulk ships, the default utilisation factor of 0.7 was kept unchanged, since cargo ships tend not to cover the return trip empty; this value agrees with the estimates reported by the IMO [51]. For road transportation, the GaBi default utilisation factor is 0.61, meaning that trucks travel with an average of 61% of its full capacity (by weight). However, trucks in the analysis are always considered to return empty, which lowers the maximum utilisation factor to 0.5. It was thus decided to rely on Ecoinvent data, which for trucks with a payload of 18-32 tonnes set this parameter at 0.48 [108]. For rail cargo transport, the GaBi default utilisation factor of 0.56 for bulk goods was adopted.

Emission standards in Israel, US and Europe are similar, and the study considers trucks under Euro4 compliance in all locations. In a conservative optic, the share of biogenic carbon in fuels was set at 5% for Europe and null for Israel and the US, although diesel blending rates may be slightly higher in all three contexts.

The share of road categories - rural, motorway and urban - was not changed from default values. Sulphur content does not affect the impact category of climate change and was thus kept unchanged.

In general, land travel distances were estimated using road and rail maps, when the exact location of the start and end points were known. The location of the two ginning facilities in Israeli territory is known; the model is shaped after the land route leading from the Northern gin - shown in figure 4.12 - to the port city of Haifa, in the North of the country.

From there, sea distances were estimated using official sea routes; besides Haifa port for Israel, New York port was considered for feed shipped from the US, Gela port for imports of oil to Italy, and Constantza port in Romania for export of grains to Israel; all distances included the return trip. Trains were always supposed to be powered by electricity, supplied by the average EU grid for European trains and by the average East US grid for American trains.

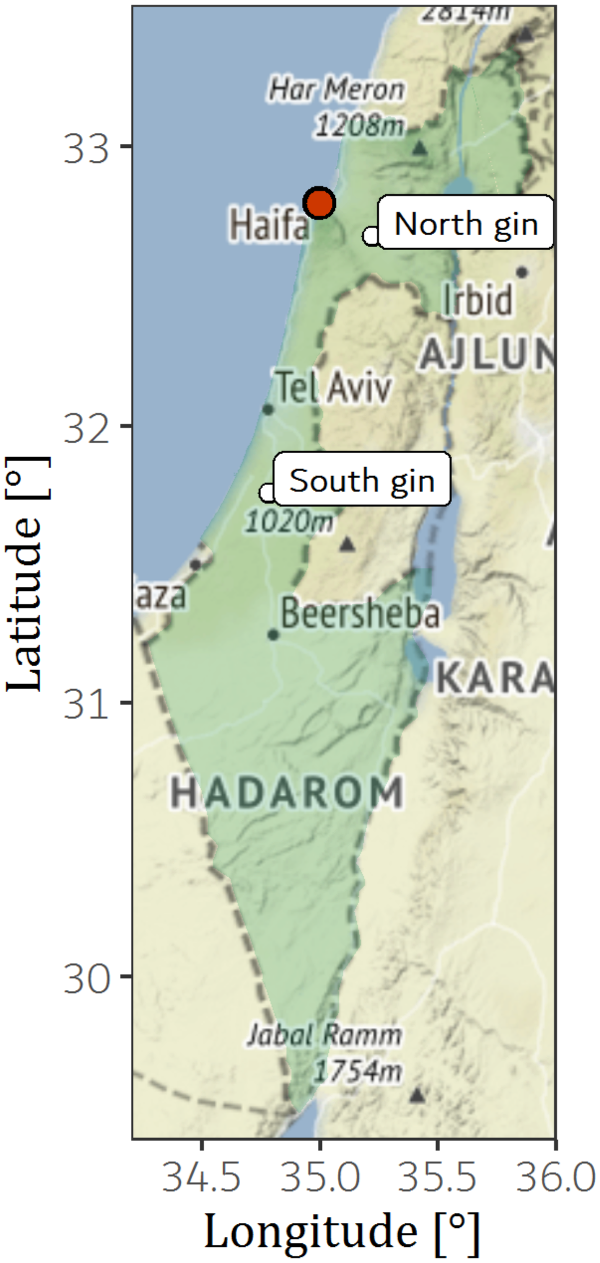

Figure 4.12: Location of the two ginning facilities in Israel (white dots), and of the city of Haifa (red dot).

4.3.6 Electricity supply

For each location under analysis, electricity supply is modelled as coming from the average electricity mix. Matching the electricity demand for each unit process with time-dependent electricity mixes would have been too hard, if not impossible, and not in the scope of the present research.

The investigated product system involves electricity grids in three locations: the EU, the US and Israel. In the EU, the average electricity grid mix was used for train transport as well as for grains production; electricity used in biorefining was instead modelled as coming from the Italian electricity grid. Processes taking place in the US rely on the electricity grid model for Eastern US. All the aforementioned grid supply modules were available in the GaBi database at disposal.

Since database information did not include any module for Israel, the Israeli electricity grid had to be entirely modelled. Data on electricity production were first sourced from the IEA database, for year 2017 [143]. For national security reasons, Israel grid is isolated: consequently, the production mixes of neighbouring countries do not influence its electricity emission intensity. The production shares, as presented in figure 4.13, favour fossil fuels, with renewables constituting less than 3% of the production; within renewables, PV represents more than two thirds of the production, the remaining being split between wind power and a generic category "other sources". Natural gas use is dominant, since the country has abundant gas reserves and limited export possibilities of such gas. The remarkable presence of coal, in particular, makes the Israeli grid more carbon-intensive than the average European grid.

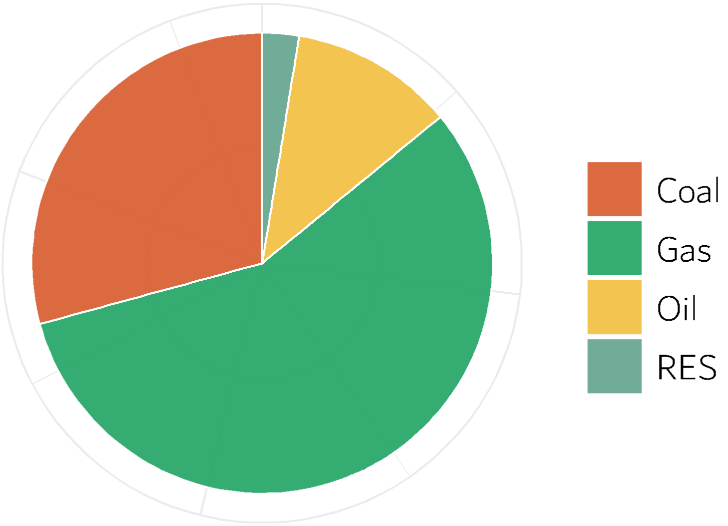

Figure 4.13: Domestic electricity production shares by source for Israel in 2017.

Since Israel does not make use of hydroelectric, it was deemed as sufficiently accurate to rely on data from a single year. The power supply was modelled after EU plants, using the GaBi database, since it was not possible nor within the scope of the analysis to model each specific source. The category "other", covering 242 GWh_{el}, was considered to be constituted entirely of electricity from concentrated solar power, since it is likely that it is mostly covered by the 121 MW Ashalim solar tower in the Negev desert [144]. The supply model finally included energy industry own use (about 4.5% of gross production in 2017) and transmission losses (about 3.5%).

4.3.7 Modelling of Indirect Land Use Change effects

Additionally to cultivation emissions, the supply of animal feeds implicate an ILUC effect that cannot be omitted when assessing the sustainability of the life cycle under examination: even though cottonseed itself has no ILUC implication in our system, the supply of cereals does have an ILUC effect which ought to be attributed to cottonseed-oil RD.

There are three ways in which an increase in demand for an agricultural good - in this case feed grains - may be balanced by the global market: by changes in demand, in yield and in cropped surface. In particular, the increased demand for animal feeds induced by cottonseed oil HVO in the form of reduced supply of cottonseed to the dairy industry may interact with the global market in three ways:

- Consumer demand may decrease, owing to the higher price of grains.
- Increased demand may spur, in the farmers' interests, technological advancements to increase grain yield.
- Increased demand may cause some conversion of land into new agricultural land.

The third of these mechanisms corresponds to ILUC. Its effects are so uncertain that they are normally reported separately from other emissions, a logic followed by this study as well. The aim is not to assess with precision GWI values including ILUC, but rather to see if this phenomenon is expected to increase or decrease the GWI difference with other oil feedstocks.

Due to its global, indirect nature, ILUC cannot be observed nor measured, and its estimates must rely on models. Many different approaches exist, with the two most notable types based on partial equilibrium or general equilibrium models; these can both have either a local or a global perspective, with general equilibrium models being able to capture more economic feedback effects than partial equilibrium models, but also being very sensitive to their parametrisation. The most well-known general equilibrium models for ILUC are GTAP and MIRAGE - the latter adopted as reference by the European Commission for its policy decisions.

Models study the impact of crops directly used in biofuels production, and their values are normally reported as CO_2 per unit of ethanol LHV produced; nonetheless, their results can be adapted to describe increased feed demand as a consequence of biofuels production, and thus be used in the present study as well. Although covering the whole global economy, GTAP and MIRAGE work based on a demand shock for biofuels which has a local nature - GTAP studies focus on demand in the US and MIRAGE studies on the EU. No study has ever been so specific as to investigate ILUC effects of an increase in Israeli cottonseed oil demand from the EU, so that estimates have to be borrowed from general research. In this respect, any ILUC effect caused by augmented demand for grain feeds in Israel is not likely to have effects on local production, since agricultural land is rare and thus highly valuable in this country. Grains would instead spur imports from a range of partner countries, like the already mentioned Ukraine, EU, US. Notwithstanding, the main uncertainties in ILUC are to be found in model parametrisation and assumptions, and the issue of the location of demand shock and grain production is secondary to these other issues.

The analysis of ILUC effects opened with a literature review. In this respect, the first observation is that, after the first publications in the late 2000's and up to year 2012, ILUC estimates across different models have shown a gradual trend of convergence towards lower values than originally estimated [145]; since then, little progress has been done in further reducing the uncertainties surrounding ILUC values.

The literature review focused on the two most relevant general equilibrium models, MIRAGE and GTAP. The various estimates that have been published over the years are collected in figure 4.14: all values reported in this section are computed by distributing the total soil carbon depletion due to LUC in 20 years, coherently with EU regulation [7]; in some cases, studies are based on a 30-year amortisation period, most notably American studies, so their value has been recalculated. [8] The next paragraphs will start by discussing each of the two model and its progress in estimating ILUC.

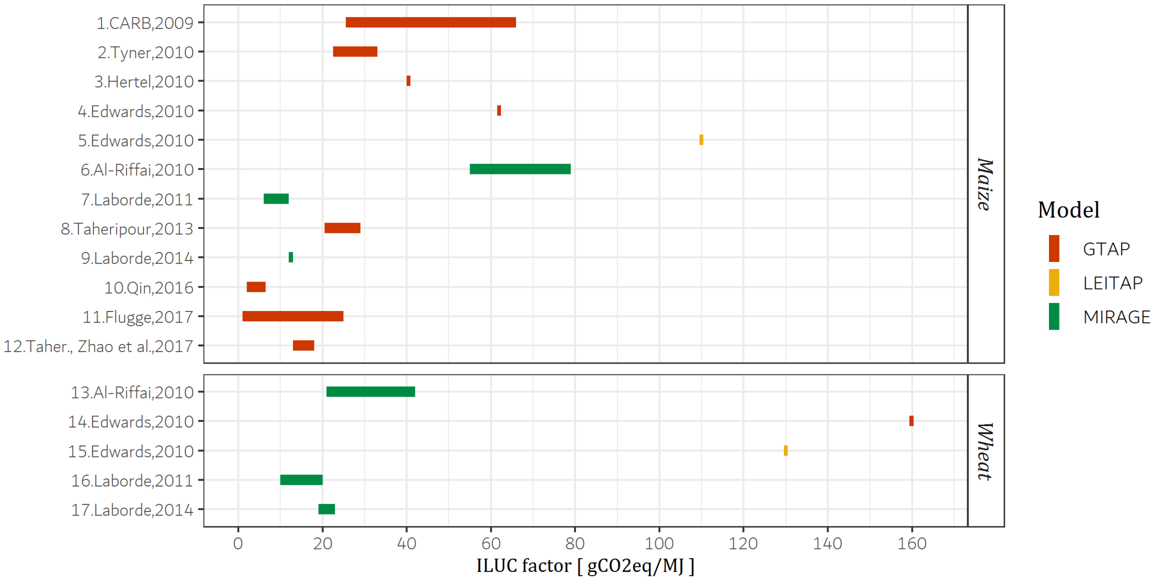

Figure 4.14: Review of ILUC emission estimates of maize and wheat in past literature (emissions amortised over 20 years) [145] [146] [147] [149].

Although disagreement on the magnitude of ILUC effects is high, some important common features between different studies can be traced: firstly, studies usually tend to show less result variations in their estimates for cereal-based ethanol than in their oilseed-based biodiesel estimates; secondly, studies agree that cereals induce less ILUC

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[8] ILUC emissions depend mainly on the change in soil carbon stock: when forestland is turned into cropland, the forest carbon-rich soil starts releasing stocked carbon to the carbon cycle. In the accounting of ILUC emissions in form of a coefficient, this carbon must be amortised over a time frame. US studies and regulations usually adopt 30 years, whereas in the EU the usual time is 20 years.

emissions than conventional oil crops: this is an important conclusion, whose consequences will be analysed in section 5.2.6 of the results.

MIRAGE is the model used to inform European policy makers of ILUC effects; there has been three major versions of MIRAGE models and results from 2010 to 2014; each new version served as refinement to the previous one. The last review was carried out by Laborde in 2012-2013 and its results for cereals ethanol stabilised around 12 $^{g_{CO2}}/_{MJ}$, with sensitivity analysis resulting in a range of 7.6 to 16.5 $^{g_{CO2}}/_{MJ}$ [93]; the study provides also crop-specific values for maize and wheat ethanol.

GTAP is a model developed in the USA by researchers at Purdue University, and it features a larger number of publications compared to MIRAGE. As it can be seen from figure 4.14, the first studies using GTAP generally reported higher estimates than later work on the same model; successive works from 2013 onwards show a growing convergence towards lower results, with estimates as low as about 2 $^{g_{CO2}}/_{MJ}$ [146] [147]. However, some of the main assumptions at the base of these researches have been questioned in recent publications. In particular, criticism mounted over the choice of yield-to-price elasticity, a key parameter in GTAP models: in new models, this value is generally increased without solid empirical evidence; accompanied by higher cropping intensity, i.e. higher number of harvests per year on the same area, this value is said to reduce ILUC emission factors of about 35% [148]. Other poorly-supported hypotheses like the regionalisation of said elasticity index, emission factor oddities for certain land types and estimates of expansion into peatland for certain crops caused a general and severe reduction in ILUC results from all GTAP studies after 2012 [148] [149].

Based on this analysis, choosing the latest studies available does not seem to guarantee any more accuracy than older estimates; it was thus chosen to rely on the latest MIRAGE estimates available, from the already mentioned (Laborde, 2014); this same study was the one used for reference values of ILUC emissions in EU legislation amending the RED, published in 2015 [150]. It is important to underline that the level of precision of ILUC estimates, in general and in the framework of our study as well, may just allow for a cautious comparison of the analysed pathway with other competing pathways.

Coherently with EU standards and common practice, ILUC emissions will be presented separately from all others emissions; as cottonseed oil HVO causes ILUC effects not by itself, but by inducing increased demand of maize, wheat and barley, such ILUC effects will be compared with other common HVO feedstocks. The scenario "STEP1" combining *2020 yields corrected* and *No "other oilseeds" to arable in EU* is considered to be the best starting point for the present analysis [93, p. 26]. The objective, at this point, is two-fold: (I) find an ILUC emission factor for cottonseed-oil RD; (II) find ILUC emission factors that appeal to palm and rapeseed oil RD, with the purpose of making a comparison with cottonseed oil possible. The latter point will be tackled first, since it follows a more straightforward logic. The underlying assumption in grains ILUC emissions estimation is that their effect can be related to grains used in the bioethanol industry, for which emission factors are available in $^{g_{CO2eq}}/_{MJ}$ of ethanol.

It was first necessary to refer these ILUC emissions to RD fuel by first relating these emissions to the unit mass - of grain or oilseed - upstream of the biorefinery: for this purpose, conversion efficiencies and fuel properties had to be assumed based on standard practice; the same was done for cottonseed and the other feedstocks for which a comparison is to be made (palm and rapeseed) [18] [7] [93]. The transformation is carried out by using the following two factors: 96% for mass conversion of vegetable oil to biodiesel (FAME), 34% for mass conversion of maize to ethanol and 36% for mass conversion of wheat to ethanol. The sources for these values are reported in the table by equations 4.16a and 4.16b, showing how the computations were carried out.

$$EF_{oil} = EF_{FAME} * LHV_{FAME} * CF_{FAME} \tag{4.16a}$$

$$EF_{grain,i} = EF_{eth,i} * LHV_{eth} * CF_{eth,i} \tag{4.16b}$$

EF_{oil}	ILUC emission factor of palm/rapeseed oil	g_{CO2eq}/kg	2001	calc.
EF_{wheat}	ILUC emission factor of maize	g_{CO2eq}/kg_{dry}	118	calc.
EF_{maize}	ILUC emission factor of wheat	g_{CO2eq}/kg_{dry}	174	calc.
EF_{FAME}	ILUC emission factor of biodiesel	g_{CO2eq}/MJ_{bd}	55	[93]
LHV_{FAME}	Default biodiesel lower heating value	MJ/kg	38	[7]
CF_{FAME}	Conversion factor biodiesel-vegetable oil	kg_{FAME}/kg_{oil}	0.96	[18]
$EF_{eth,maize}$	ILUC emission factor of maize ethanol	g_{CO2eq}/MJ_{eth}	12	[93]
$EF_{eth,wheat}$	ILUC emission factor of wheat ethanol	g_{CO2eq}/MJ_{eth}	19	[93]
LHV_{eth}	Default ethanol lower heating value	MJ/kg	27	[7]
$CF_{eth,maize}$	Conversion factor ethanol-maize grains	kg_{eth}/kg_{dry}	0.364	[18]
$CF_{eth,wheat}$	Conversion factor ethanol-wheat grains	kg_{eth}/kg_{dry}	0.339	[18]

After reporting ILUC emission factors per unit of feed upstream of biorefining, a conversion factor from grain to RD fuel has to be introduced: it was decided to rely on the conversion factor CF_{RD} proper of the baseline case of the present analysis, i.e. 74.4 kg_{RD}/kg_{oil} (in short, the baseline diesel yield). The result is 2.72 kg_{CO2eq}/kg_{RD}

The emissions per unit of feed had then to be allocated to the specific hydrogenated product of interest, namely RD. Using energy allocation and the same product yields - of diesel, naphtha and light products - as in the baseline of this study, it is found that RD is assigned with about 82.5% of total emissions, i.e. with 2.23 kg_{CO2eq}/kg_{RD}. [9] Translated per unit of energy, this emission factor becomes 51.8 g_{CO2eq}/MJ_{RD} for both palm and rapeseed.

For grains, it is in principle sufficient to base the calculation on the quantities consumed per MJ of diesel produced in the respective scenario; however an important correction

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[9] The same procedure, in principle, may be applied when passing from ILUC emissions per unit mass of biodiesel to emissions per unit feed, since transesterification delivers about 100 kg of glycerine per ton of feed. However, all emissions are applied to biodiesel, since glycerine, which is currently in high excess in global market, is more of a waste than a co-product.

must first be made to EF_{maize} and EF_{wheat}. It is in fact necessary to account for the lack of co-products in the use of wholegrain, as opposed to the grain-to-ethanol pathway, which generates considerable amounts of distillers' dried grains; this protein feed prompts for some avoided animal feed, thus mitigating the detrimental effects of ILUC. This effect must be removed, as the current research does not feature any such co-product: ILUC happens because of the increased demand for whole grains by the animal industry. The effects of such co-products was removed based on an alternative scenario from the same publication on MIRAGE estimates, called *no co-products* scenario. The purpose of this scenario is exactly to test this effect. Here, ethanol maize is registered to see its ILUC coefficient increase by +46%, and wheat by +28% [93, p.32]. Thus, the respective emission factors can be corrected to 172 g_{CO2eq}/kg_{dry} and 223 g_{CO2eq}/kg_{dry}. The underlying assumption is that the interactions between the *no co-products* scenario settings and the settings of or main scenario are negligible, so that these increases can be transposed as they are.

It is worth mentioning that other studies may lead to much different outcomes. A study using GTAP to estimate ILUC emissions of US maize ethanol found a much more consistent increase in total agricultural output requirement after elimination of co-products, from +36% to +17%. If this translated entirely in land use change, it would correspond to an increase of grain cropped surface by 110%. In any case, results are hardly comparable, since the values from this study are reported only in terms of additional agricultural output, without specifying what crops would expand, nor if agricultural yield increases play any role in this output increase. Nonetheless, a European study on barley used this source for the evaluation of barley ILUC emission factor [122].

It is now sufficient to account for the quantities of each feed in the feed schemes considered to obtain ILUC emission estimates for a given scenario, as reported in section 5.2.6 of the results.

Results, critical analysis and conclusions

5.1 Life Cycle Impact Assessment

This LCA deals with the sustainability of cottonseed-oil RD under a greenhouse-gas (GHG) emissions perspective. The impact category investigated is *impact on climate change*, through the use of the category indicator *radiative forcing*, which is the physical phenomenon linking GHG emissions with climate change. The characterisation factor utilised is *Global Warming Potential (GWP) 100 years*, expressed in $^{g_{CO2eq}}/_{MJ}$. In this analysis, such characterisation factor does not account for biogenic carbon, i.e. carbon of immediate biological origin. For this reason, the carbon uptake at plant growth is not accounted for, and neither is the carbon release as CO_2 at combustion of the biofuel, when included in the system boundary; the same holds for emissions of water vapour, which, in principle, is also a GHG. The analysis results in a value of Global Warming Intensity (GWI) of the RD fuel, expressed as well in $^{g_{CO2eq}}/_{MJ}$. The difference between the terms GWP and GWI is subtle: while GWP is used to refer to greenhouse gases and their induced global warming, GWI refers to the greenhouse effect related to product flows.

The GHGs impacting on the assessment are carbon dioxide, methane, nitrous oxide; These are assigned characterisation factors in accordance with IPCC guidelines and the requirements of EU legislation, as reported in table 5.1. The category endpoint is the atmosphere, where greenhouse gases end up.

All model results have been computed following the framework described above. The results for the baseline of models EUC and IS are reported in figure 5.1: the level of aggregation presented here allows to distinguish the different forms of transport of cottonseed and related sub-products; it is also possible to distinguish the different phases of processing.

The EUC model results in a GWI of 15.7 $^{g_{CO2eq}}/_{MJ}$ of RD, while the IS model stands at 83.7 $^{g_{CO2eq}}/_{MJ}$. Coherently with its definition of agricultural residue in EU legislation, cotton cultivation and ginning do not contribute to this biofuel GWI in the EUC model.

Table 5.1: Set of greenhouse gases considered in the present analysis with their relative characterisation factor, i.e. GWP 100 years $[kg_{CO2eq}/kg]$. Values coherent with European regulations and IPCC AR4

Greenhouse gas	**Characterisation factor (GWP)**
CO_2	1
CH_4	25
N_2O	298

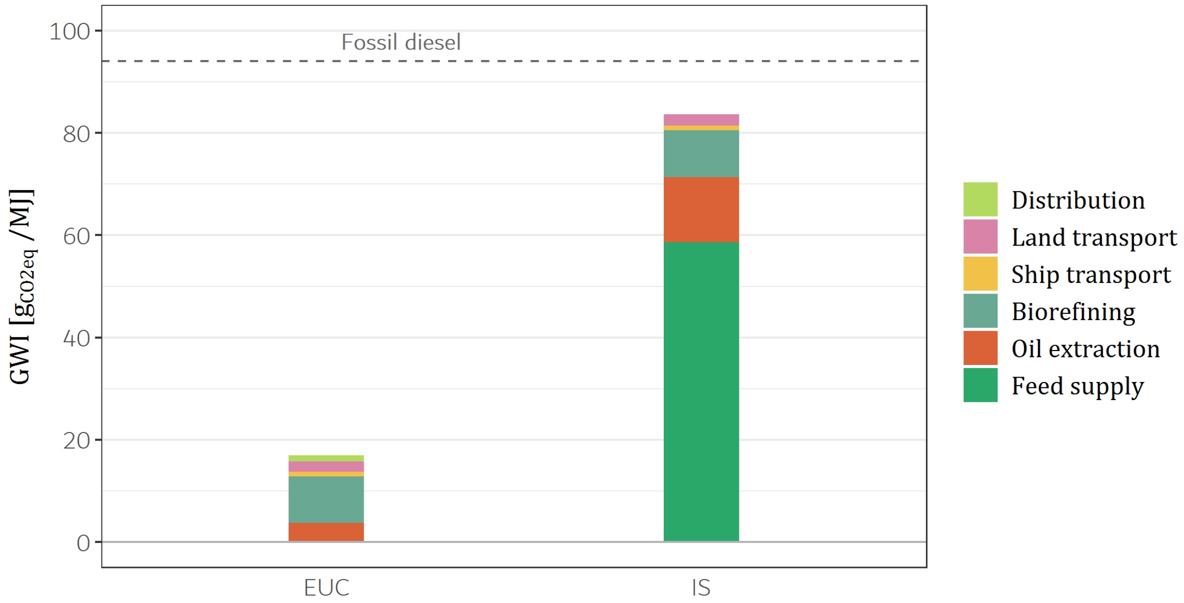

Figure 5.1: GWI baseline results in the EUC and IS models.

The net contribution is also null in the IS model, regardless of the definition of cottonseed, since the system boundaries allow to consider a replacement of cottonseed through cottonseed meal and feed supplements. These supplements - US maize and wheat in the IS baseline (BL) feed supplement scheme - represent the main contribution to GHG emissions in this model, making the biofuel consistently more emission-intensive than what results from the EUC model: in fact, feed supply alone represent about 70% of the total GWI of the biofuel. Note that *feed supply*, here, entails emissions starting from the inputs for the cultivation of grains and up to delivery of such grains to the dairy farm, including all transport processes in-between; *Land transport* and *ship transport* as reported here, on the other hand, are related to cottonseed and all its sub-products. The oil extraction phase contributes to further enlarging the GWI difference between the models, due to the presence of energy allocation at the level of its outputs in the EUC model, which lowers the GHG emission intensity of cottonseed oil in this phase by a factor of around 3.5. The EUC model entails also a distribution process, as requested by regulations: the related GWI component is taken from the default values of the directive REDII; the IS model, on the other hand, stops at the level of biorefinery outputs, excluding distribution.

Figure 5.2 offers a breakdown of the two processing stages, oil extraction and biorefining. The former features emissions from electricity and heat demands in the proportion of one and two thirds respectively; a minor contribution of around 1% is linked to hexane supply. [1] Biorefining stage GHG emissions are instead dominated by hydrogen supply through steam reforming, at around 90% share of GWI; the remaining emissions are divided between electricity and steam supply.

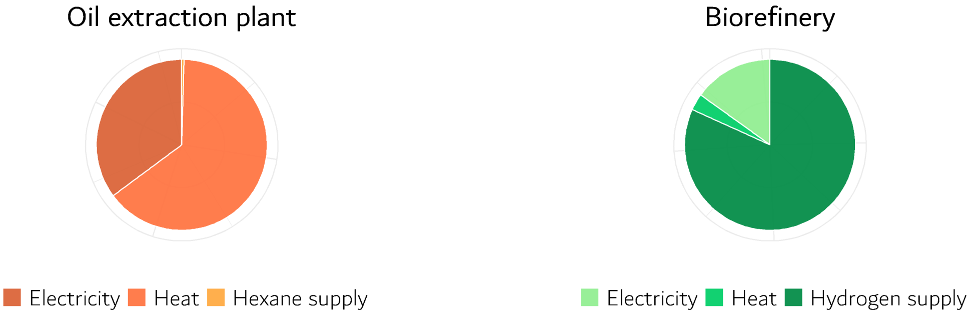

Figure 5.2: Breakdown of GHG emissions in the oil extraction stage (left) and in the biorefinery stage (right).

These two baseline scenarios have been subjected to a change in all the main uncertain parameters, in order to estimate a range of variability for their GWI. As presented through section 4.3, some inventory information are intrinsically uncertain; each of these parameters has thus been assigned with a range of variation which, in the absence of enough information to develop probability distributions, have been grouped according to their impact on the final GWI value: consequently, the baseline scenario can be compared with a Low-CO2-Equivalent (LO) scenario and a High-CO2-Equivalent (HI) scenario; table 5.2 shows the set of parameters that were tested. Note that the only system component where parameters are left unchanged is feed supply. The range of variation for each parameter is explained in the relative part of section 4.3.

This analysis delivered the results of figure 5.3. First, it can be noticed how the most affected processes are oil extraction and, in the IS model, feed supply, while transport processes and biorefining change only slightly from one parameter setting to the other. The first peculiarity is the remarkable difference in range of variation depending on the model: in EUC, the GWI varies only by about ±5%, whereas the IS model feature a variation of about ±15%. As thoroughly illustrated in the next section, the parameters of oil_{dry}, Y_{extr}, $mc_{extr,in}$ and St_{extr} all contribute synergistically to a decrease or increase in impact of the extraction plant; while energy allocation downstream of this unit in the EUC model allows to distribute this change among the co-products of oil and meal, in the IS model all the burden remains within the product system.

Moreover, the system expansion applied in the IS model implies that a change in

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[1] In accordance with conventions and EU requirements, only direct radiative forcing is considered; consequently, hexane emissions to air from the extraction plant do not account for any increase in GWI, even though such volatile paraffins cause some indirect radiative forcing [151].

Table 5.2: Parameters included in the scenario impact analysis (BL=Baseline; LO=Low CO_{2eq}; HI=High CO_{2eq}).

Parameter	**Explanation**	**LO**	**BL**	**HI**
oil_{dry}	Oil content in dry cottonseed	0.22	0.18	0.16
Y_{extr}	Oil extraction yield	99%	95%	90%
$mc_{extr,in}$	Moisture content in seed entering the extraction plant	8%	10%	12%
St_{extr}	Steam consumption factor at the extraction plant	75%	100%	125%
Ut_{bioref}	Utility consumption factor	100%	100%	110%
r_{dehull}	Load ratio at dehuller	116.5%	100%	83.5%

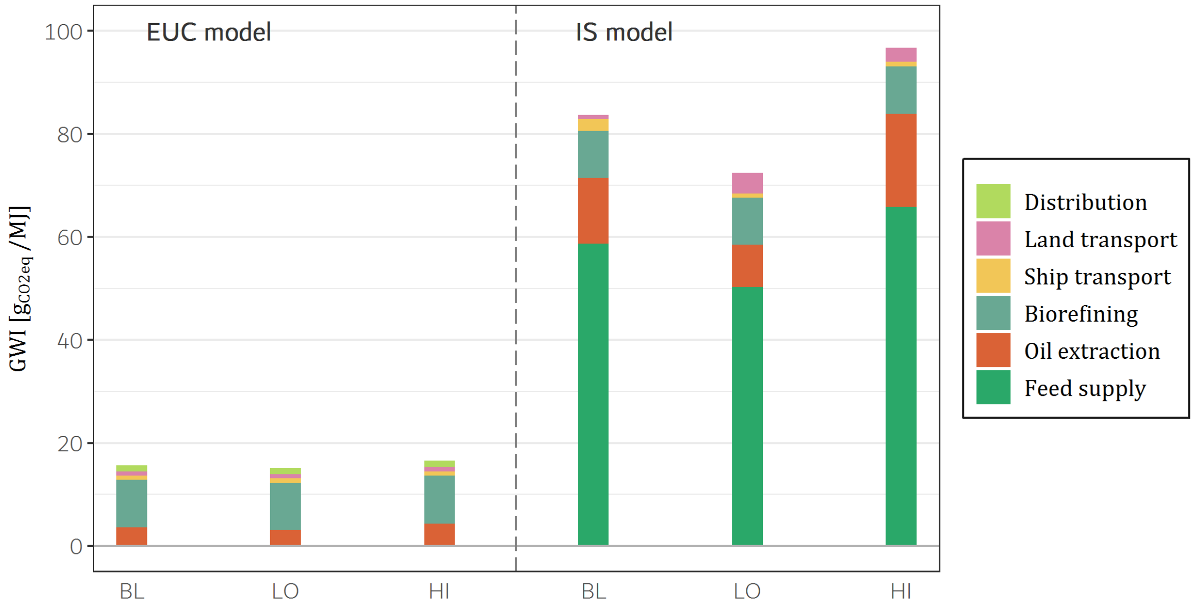

Figure 5.3: Global warming intensity breakdown for EUC and IS model under different parameter values, capturing its range of variation.

nutritional properties or quantities of meal impacts on feed supplement requirements: both a lower seed oil content and a lower oil yield - as further explained in section 5.2.3 - bring about higher feed supplement needs, which happens to be the most relevant source of emissions.

From here on, the presented range of variation will be shown as whiskers in the plots. In the next section, results will be further analysed in the framework of the research goals, discussed and compared with other competing HVO pathways.

5.2 Interpretation of the results

5.2.1 Compliance with EU emission savings requirements

By following the methodology imposed in EU legislation through the directive known as RED II, the EUC model and its range of variation allow to assess the compliance of cottonseed oil with European directives in matter of biofuels. According to RED II, biofuels production plants which entered operation in-between October 2015 and December 2020 must comply with a 60% GHG emissions reductions over their fossil counterpart; the fossil fuel counterpart of renewable diesel (RD) being fossil diesel, which is standardised at a GWI of 94 ${}^{gCO2eq}/_{MJ}$, such GWI threshold stands at 37.6 ${}^{gCO2eq}/_{MJ}$.

Cottonseed-oil RD is assessed at a Global Warming Intensity of around 16 ${}^{gCO2eq}/_{MJ}$, thus offering a GHG emissions reduction of more than 80%. Furthermore, the range of variation, as assessed in this research, is contained at around ±5%. For this reason, this outcome is judged as solid and reasonably applicable to other contexts similar to our case study without important turnovers, making cottonseed-oil HVO a feasible feedstock option for European biorefiners. Even if the GWI were twice as much as the one assessed in this research, the fuel would comply with regulations.

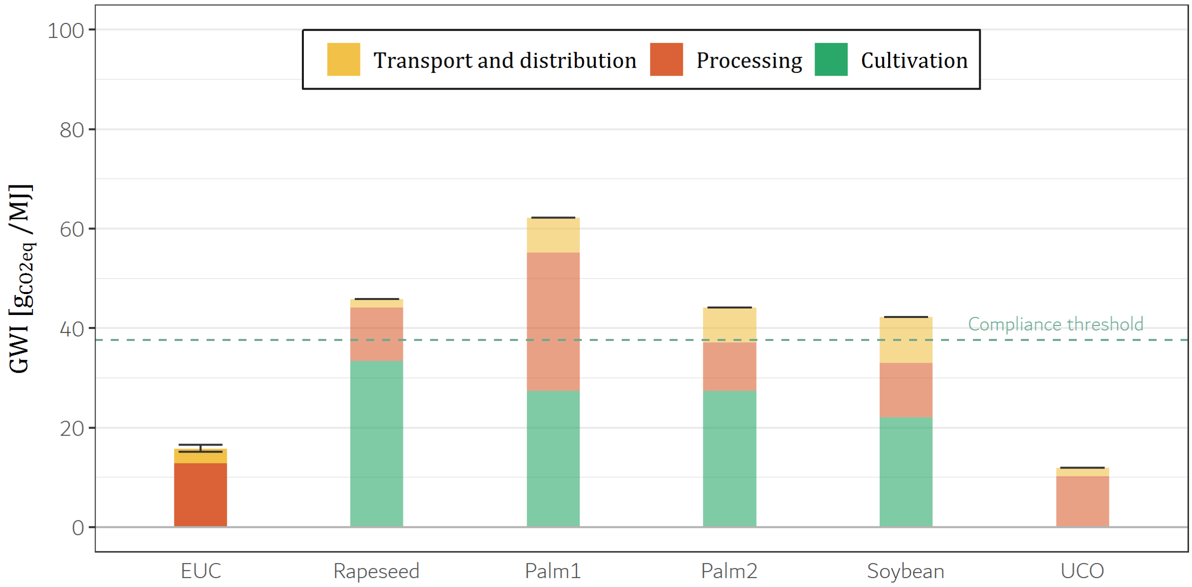

Figure 5.4: Global warming intensity of renewable diesel as from the EUC model, compared with other competing feedstock options (Palm1=palm oil without CH_4 capture; Palm2=palm oil with CH_4 capture; UCO=Used Cooking Oil).

Figure 5.4 shows a comparison between the EUC model and other common RD feedstocks. All the values used in the comparison are reported in the EU directive RED II as "typical GWIs" for RD production, and thus use the same exact methodology. In this graph, it is possible to appreciate how cottonseed-oil RD is second only to used cooking oil (UCO) in terms of GWI. It can also be seen how, considering the typical value, the usual first-generation feedstocks fall short of GHG reduction requirements. Producers must thus actively demonstrate the compliance of their fuels by means of a voluntary scheme, a recognised form of certification that attests the environmental performance of a biofuel. As expected, the largest contributor to most GWI values tends to be the agricultural phase, which is totally absent in the EUC model thanks to the definition of cottonseed as agricultural residue.

5.2.2 The importance of feed supplements

In the IS model, the most relevant share of GHG emissions originate in the supply chain of grain feeds, which are consumed in the dairy farm to supplement cottonseed meal; the IS baseline (BL) case features US maize and US wheat as feed supplements and the GWI of their supply represents about 70% of the total biofuel GWI value. Since the US and the EU are very close as suppliers of grains to Israel, it is important to test alternative grain supplies from the EU. As already mentioned, two alternative *feed supply schemes* have been investigated: BDK, which features barley from Denmark, and WEU, which features wheat from an average of EU production. The inventory data and the procedure to estimate the GWI of each grain are explained in section 4.3.3.

Figure 5.5 shows the final results for each of the three feed supply schemes, comparing these results with competing feedstock options as from a recent LCA study on RD [14]. The GWIs of these other fuels have been assessed using the same methodology as in the IS model: in these cases, seed meals are set to displace soybean meal - which is usually regarded as the marginal protein meal - while biorefinery products are subject to energy allocation. Note that there is a discrepancy between our study and the one just mentioned, given by the use of a characterisation factor for N_2O of 320 instead of 298; this implies a slight overestimation of field emissions impact compared to our case.

The figure shows that nearly 20 $^{g_{CO_{2eq}}}/_{MJ}$ separate the BL from the WEU scheme. Thus, in terms of comparative analysis, changing feed supply from American maize and wheat to EU wheat would make the produced RD less GHG intensive than rapeseed and jatropha biofuels.

A first reason for this difference is that European feeds are more efficient, in terms of GHG emissions, than the American feeds featured in the BL case. Extensive quantities, in terms of grain mass, do not differ appreciably from feed supply scheme to feed supply scheme: in all cases, per kg of displaced cottonseed, roughly 50 g of grains are necessary; barley is required at the amount of 54.8 g, wheat in WEU at 51.8 g, maize and wheat in BL at a total of 51.5 g, of which about 41 g are US wheat. What makes up for a different GWI is primarily the different emission intensity of cultivating these grains: as shown in figure 5.6, US wheat - by far the prevalent grain in IS baseline feed supply scheme - is the most emission-intensive cereal, especially due to its relatively low yield; the most efficient feed is instead EU wheat.

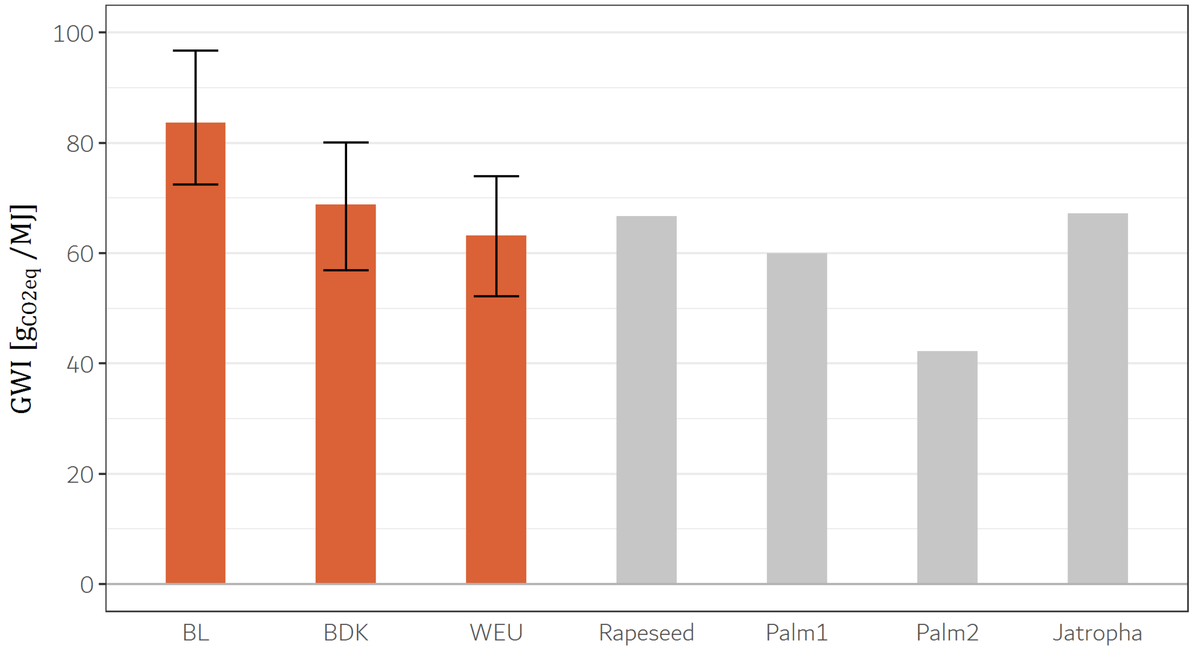

Figure 5.5: Global warming intensity of renewable diesel as from the IS model, compared with other competing feedstock options (Palm1=palm oil without CH_4 capture; Palm2=palm oil with CH_4 capture).

It is not to exclude that, by using different datasets, this difference may be reduced or even inverted.

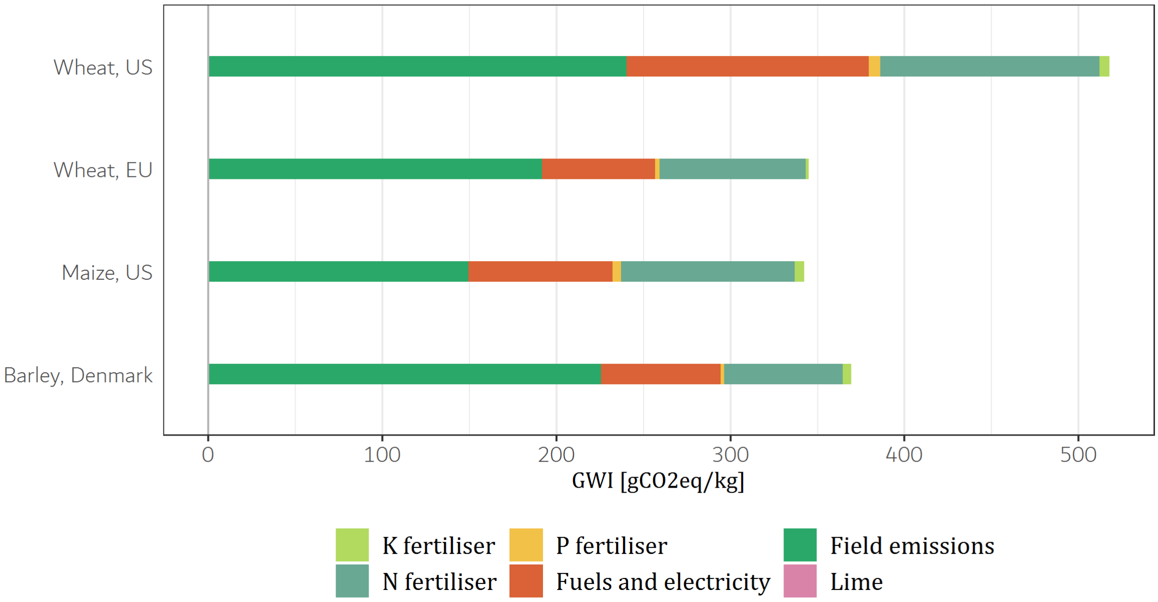

Figure 5.6: Global warming intensity of different feed supplement options modelled in this study. Quantities refer to a kg of dry grains.

A second source of divergence lies in the emissions of grain transport to Israel. Taking into account all transport emissions except the (negligible) ones linked to fertiliser supply, the baseline setting emits about 19 $^{gCO2eq}/_{MJ}$ of fuel; on the other hand, transport emissions in BDK and WEU respectively amount to 14.3 and 12.6 $^{gCO2eq}/_{MJ}$.

These results show that transport up to grain consumption location has an important effect on the biofuel GWI performance. Under the BL scheme, in fact, the supply of American grains increases the final GWI by about 16 $^{gCO2eq}/_{MJ}$, constituting about 20% of such GWI. This stresses the importance of short feed supply chains, especially for goods of relatively low density such as grains; while Israel is an especially unfavourable context for short supply chains in this sense, this principle could be applied with positive outcome in other cotton-producing countries.

The margin for a better GWI performance depends to some extent also on the electricity grid in use. The impact of the Israeli electricity grid mix, which is particularly carbon-intensive, has in fact been tested against an average EU grid, showing that the former leads to an extra 2 $^{gCO2eq}/_{MJ}$ GWI.

In conclusion, the choice of a feed supplement with low GHG intensity - with a short supply chain, using low-impact agricultural practices and delivering high yields - is particularly important for the sustainability of this fuel under the point of view of GHG emissions. In our case study, selecting EU wheat as feed leads to a GWI that is about 5% lower than rapeseed-oil RD, the most common oilseed used in the European biofuels industry.

5.2.3 Influence of oil extraction yield and seed oil content

The parameters of seed oil content and oil extraction yield are both relevant in determining the final fuel GWI.

Oil content depends mainly on environmental conditions, growth conditions and plant species; as already discussed, literature offers a rather extended range for this value. In this research, oil contents of 16 to 22% (dry matter) have been tested, with 18% as base value. An higher oil content invariably has a positive effect on the GHG performance of the system: ceteris paribus, the aforementioned oil content maximum brings about a fuel GWI of 73.6 $^{gCO2eq}/_{MJ}$; the oil content minimum, instead, brings about 90.6 $^{gCO2eq}/_{MJ}$. The main reason behind such a decrease in GWI with increasing oil content is the adjustment of feed supply: higher oil prompts less feed requirements since, ceteris paribus, the amount of seed for producing a unit of fuel decreases linearly, i.e. the amount of seed to be displaced - the dashed blue line in figure 5.7 - also linearly decreases. The GWI decreases with diminishing returns on oil content: as oil content increases, some process emissions are greatly affected - in particular of grains supply and oil extraction - whereas other processes, like biorefining, remain unchanged.

Differently from oil content, oil extraction yield brings about two competing phenomena. An increase in yield, in fact, spurs the following two effects: (I) the seed needed per unit

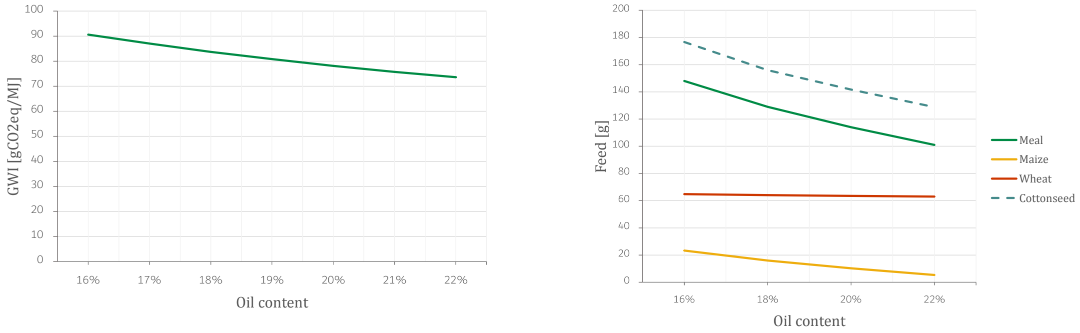

Figure 5.7: GWI depending on oil content in dry cottonseed (left) and feed supplement requirements varying the oil content (right), in dry matter, per MJ of RD. Note that the dashed line for cottonseed flow indicates a negative flow.

of fuel decreases; (II) the caloric properties of a unit mass of meal decrease. Just like with oil content, the first effect is beneficial for GWI; the second effect alone, instead, would bring to higher feed requirements. Figure 5.8 shows system behaviour under such circumstances.

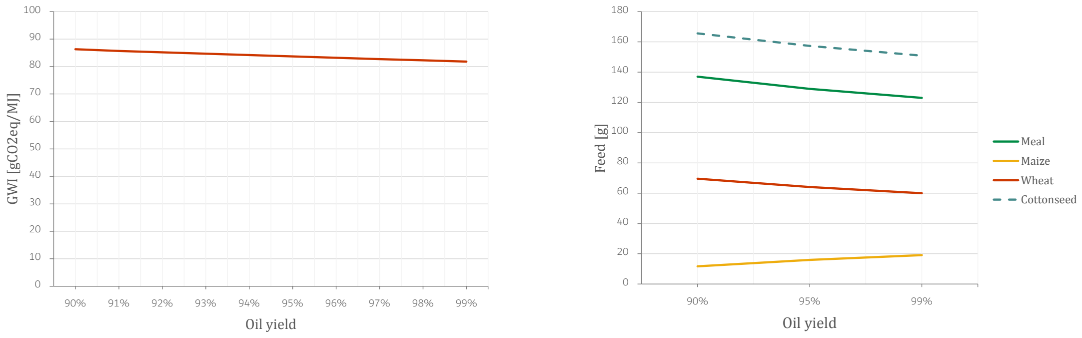

Figure 5.8: GWI depending on oil yield (left) and feed supplement requirements varying the oil content (right), in dry matter, per MJ of RD. Note that the dashed line for cottonseed flow indicates a negative flow.

Total feed requirements, net of these two contrasting effects, is observed to decrease with oil yield, so that the effect of yield on GWI is beneficial. Although less evident than with oil content, this trend also features diminishing returns with increasing oil yield for the same reason as above. This can be seen more explicitly in figure 5.10 at the end of the section, showing a large interval of 60 to 99% oil yield to prove this point. In the BL feed scheme, quantities of the two feeds relative to each other change with changing oil yield, with wheat leaving a larger share to maize as oil yield increases. Here, this has a beneficial effect as well, since maize supply is more GHG-efficient than US wheat supply.

In synthesis, the model shows that oil content potentially exerts more influence on the GWI than extraction yield. While one extra percentage point of oil yield in the range 16-22% brings about a change in GWI of -2.8 $^{g_{CO2eq}}/_{MJ}$ on average, one extra point of oil yield in the range 90-99% causes an average change of -0.51 $^{g_{CO2eq}}/_{MJ}$; at the same time, while extraction yield is linked with process engineering, seed oil content is much more difficult to control.

These parameters have also been tested for the alternative feed supply schemes BDK and WEU, as shown in figure 5.9 for oil extraction yield. There are no substantial trend differences with the baseline case, except for the fact that barley and wheat scenarios share very similar trends, with a lower impact of an increase in oil yield than the baseline case - ultimately caused by the different efficiencies of feed supply and by the effect of changing $(^{maize}/_{wheat})$ ratio in the BL case, which is not present in the other cases.

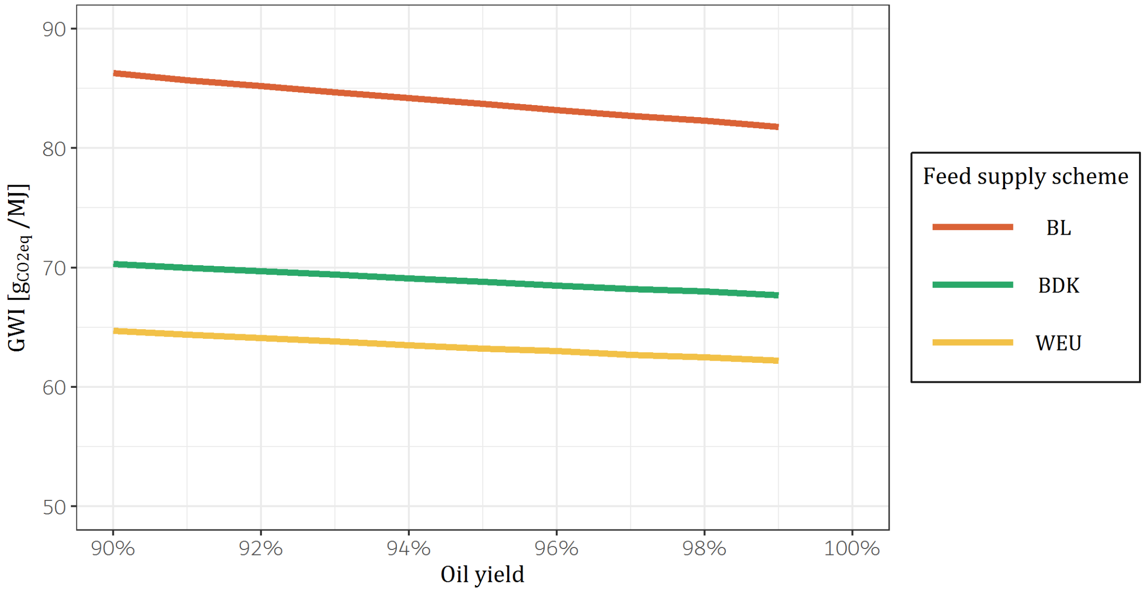

Figure 5.9: Global warming intensity of different feed supplement schemes, depending on the oil yield.

Although these results only portray the range of yields of solvent-extracted oil, the fact that all feed supply schemes feature diminishing returns implies that the marginal change in terms of GWI is more consistent at lower oil yields, as evident from figure 5.10. Thus, maximising oil yield in a situation of simple press - where yields tend to be around 60% and even lower - is more important, under a purely GHG perspective, than maximising the yield of a solvent extraction plant.

5.2.4 Influence of different biorefining setups

At the biorefinery level, two parameters in particular can influence the final GWI of its products: the tuning of hydrogenation selectivity towards the reaction of hydrodeoxygenation (HDO) or decarboxylation (DCO2), and the naphtha yield. These parameters have

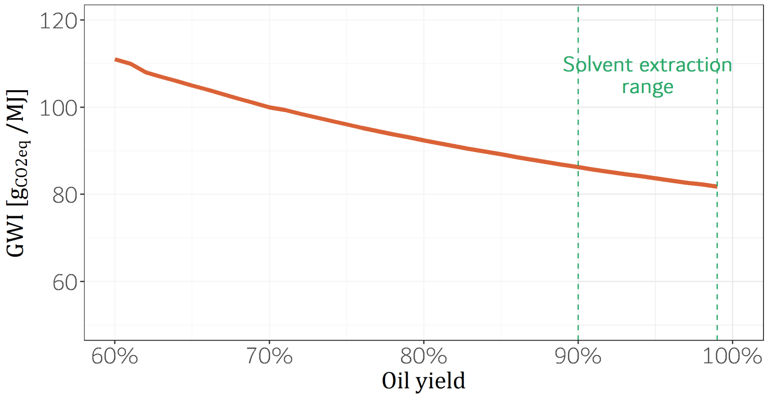

Figure 5.10: Logarithmic trend of GWI in relation to oil yield, highlighting the region typical of solvent extraction.

been tested separately, since they depend to a great extent not on variables exogenous to the biorefinery, but to the engineering of the process itself: catalyst choice, reactor temperature and residence time are the main factors explaining selectivity and naphtha yield.

Selectivity has been tested by means of a linear variation; the two extremes, i.e. selectivity purely for HDO and selectivity purely for DCO2, are presented in figure 5.11. A change in selectivity presents two contrasting effect on system performance, respectively given by a change in hydrogen demand and in diesel yield. A prevalence of DCO2, in fact, leads to lower hydrogen demand and also lower diesel yields, and vice versa for HDO; the former effect has positive consequences on GWI, since hydrogen by steam reforming is particularly impacting in terms of GHG emissions; on the other hand, the latter effect has negative consequences, since the seed needed for a unit of fuel increases. In our case, the trade-off is in favour of DCO2: the reduction in hydrogen supply, in fact, more than outsets the increase in emissions upstream of the biorefinery - linked in particular to increased feed supplement requirements and increased utilities consumption at the extraction plant. Figure 5.11 shows this effect through the two bars on the left.

This figure also highlights another consideration: the advantage of DCO2 in terms of GHG performance is higher when the system is more GHG-efficient in the extraction and feed supply sections; the two bars on the right, in fact, show the performance of a system which combines "low CO2eq" parametrisation (LO), with EU wheat feed supply (WEU), which has proved to be the least impacting in terms of GHG. In these *low impact settings*, the emission reduction from complete DCO2 rather than complete HDO is 5.6%; in absolute terms, nearly four times the reduction of a system in its baseline settings. DCO2 prompts less H_2 input at the biorefinery, but also more oil input at fixed RD output: for this reason, having a more efficient vegetable oil supply chain enhances the advantages of high DCO2 selectivity.

The effect of naphtha yield on the final GWI is much less marked: changing naphtha yield from baseline to our assumed maximum, i.e. from 7 to 13%, changes the GWI by 0.1%. This is due to the fact that naphtha yield only changes the final proportions of

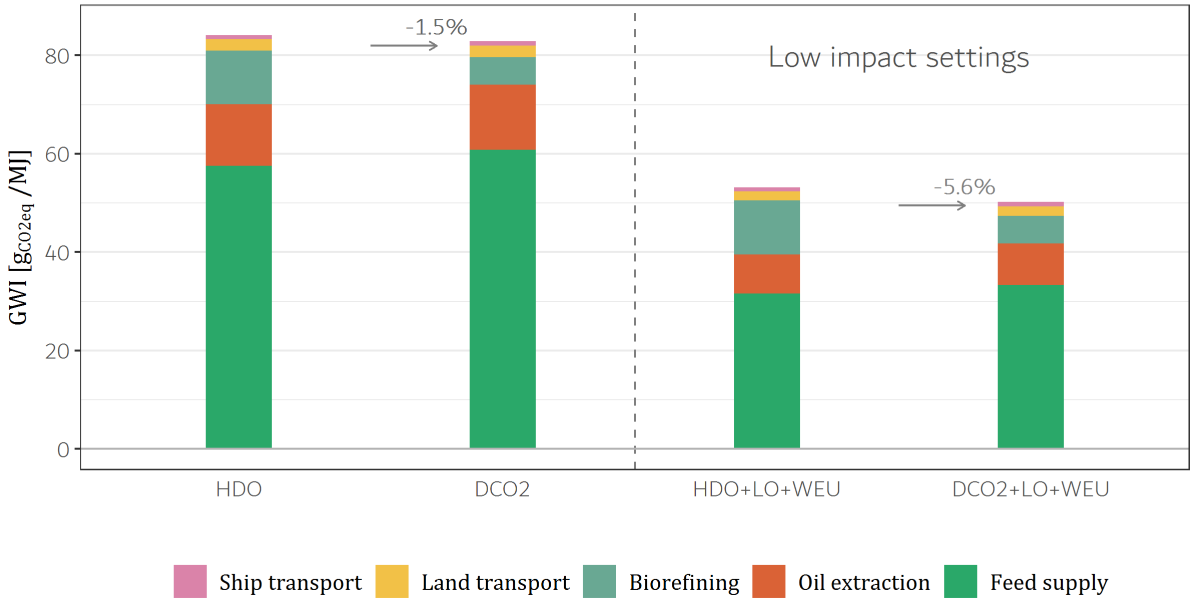

Figure 5.11: Comparison of GWI and its contributions in scenarios with pure selectivity towards either HDO or DCO2.

useful products (LPG, naphtha and diesel), which are then assigned with GHG emissions through energy allocation. Naphtha yield does not condition process consumptions nor upstream processes if not for a small increase in hydrogen consumption - as discussed in section 4.3.4. [2] Interestingly, the effect of allocation is such that higher naphtha yields subtract some GWI to RD, more than offsetting the advantage given by higher diesel yields, i.e. less cottonseed needed for a unit fuel; this is due to the higher LHV of lighter products with respect to diesel. In any case, this effect is not considered relevant on the overall results.

5.2.5 Uncertainty analysis: nitrous oxide emission factor

N_2O emissions are subject to a higher uncertainty than the factors discussed until now. The IPCC sets the emission factor for direct emissions of this gas from fields in a range spanning one order of magnitude - from 0.003 to 0.03 ${}^{kgN2O-N}/_{kgN}$, the baseline being 0.01 ${}^{kgN2O-N}/_{kgN}$. [3] Such large variation is linked to the complex nature of the phenomenon, which is influenced by crop type, soil and climate parameters, weather events, type of fertiliser, time of application. So far, all results discussed implied an emission factor of 0.01 ${}^{kgN2O-N}/_{kgN}$, which is regarded as conservative standard by the scientific community and is widely employed in academia [14] [111] [152] [153].

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[2] In principle, a higher naphtha yield is to be traced back to a higher process temperature at the isomerisation reactor: thus, it may be argued that utility consumption would increase with naphtha yield. As discussed in section 4.3.4, any additional utility consumption can be considered negligible.

[3] The unit of measurement refers to mass of nitrogen that volatilises into nitrous oxide (numerator) by the mass of nitrogen applied on fields (denominator).

An alternative to the tier 1 emission factor is the use of models, which estimate field emissions starting from information on the aforementioned factors - most importantly soil and climate; these are known as tier 2 methods. In the present study, the range of variability in emission factor from direct N_2O emissions has been tested in its impact on the results; furthermore, a tier 2 method has been applied to model IS, feed supply scheme BDK, to estimate direct N_2O emissions from fertiliser application; in this case, in fact, enough information about the location and its soil and climate conditions were available to implement the Stehfest and Bouwman model, which is also the model used in reports for the European Commission; this method is explained in depth in section 4.3.3.

Additionally to the aforementioned testing, specific N_2O emissions as reported by the datasets have been tested. In case of NREL data on maize, these emissions are calculated using IPCC tier 1 methods [125]. In case of JRC data on wheat, the method used is the combination of the Stehfest and Bouwman model with IPCC guidelines, i.e. the same as applied here for the BDK scenario, using average EU data; minor discrepancies are present in how the JRC has accounted for manure and lime emissions [18]. Lastly, in case of data on maize from (Kim,2009) and data on barley from (DCA,2016), emissions rely on tier 2 models, respectively the DAYCENT and the Daisy model [121] [122].

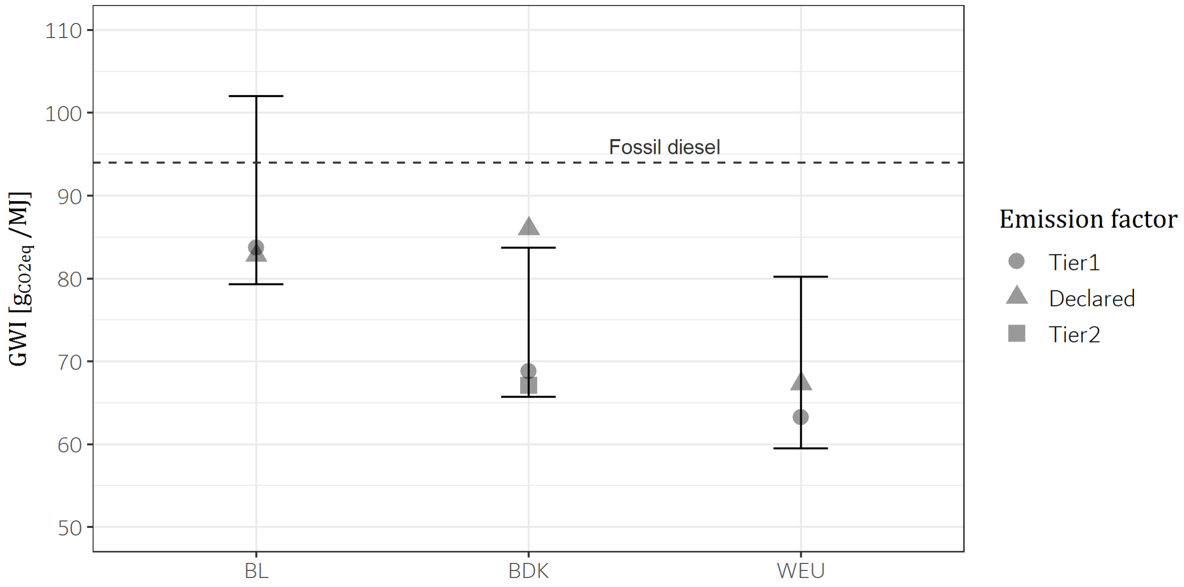


Figure 5.12: Comparison of GWI resulting from the use of different nitrous oxide emissions factors for grain field emissions.

Results are shown in figure 5.12. Under BL feed scheme, the *declared* emissions are very close to what IPCC would predict; since US wheat prevails over US maize sourced from (Kim,2009) by a factor of around 4, it is reasonable that nitrous oxide emissions are close to IPCC tier 1 estimates, which are used in the US wheat study.

In general, emission factors tend to lead to a result that is close to the tier 1 baseline; for instance, declared emissions in EU wheat lead to an increase in GWI of about 4%. However, in case of the study on Danish barley, declared emissions are much higher, and

GWI even surpasses the upper boundary set in IPCC guidelines. The study reports in fact an emissions factor of 4.2% assessed using a tier 2 approach with the Daisy model. The text supports such high value by simply stating that the model used is more advanced than IPCC guidelines [122, p.26]. The use of the Stehfest and Bouwman model for fertiliser direct emissions, on the other hand, delivers a GWI much closer to what the IPCC default emission factor would predict. All reported and calculated emission factors lead to GWIs below the fossil diesel reference of 94 $^{g_{CO2eq}}/_{MJ}$; nevertheless, what the graph points out is that N_2O emissions represent an uncertainty that is rather hard to reduce without accurate data and, possibly, field measurements.

5.2.6 Inclusion of indirect land use change effects

Up to now, all results only concerned direct life-cycle GHG emissions. Notwithstanding, the sustainability of a biofuel cannot be properly investigated without making considerations about its indirect land use change (ILUC) effects. This issue and its problematic nature has been illustrated in section 4.3.7; in the same section, an estimation of ILUC factors for HVO oilseed feedstocks as well as for the wheat and maize is carried out. Here, the aim is to compare the GWI of the IS model with other competing vegetable oil feedstocks considering the impact of ILUC. As already mentioned, this study makes use of the ILUC factors provided by (Laborde,2014) in his work for the European Commissions, values that were eventually used as reference to develop the ILUC directive and subsequent legislation [150].

This section takes into consideration the two most significant feed schemes, namely BL and WEU. BDK places in-between these two in terms of GWI. In the former, 16 g of maize and 61 g of wheat are delivered for each MJ of diesel produced; the latter requires instead 80 g of wheat. As a consequence, by applying for each grain the factors of which in section 4.3.7, it is possible to evaluate ILUC emissions at a level of 16.4 and 17.8 $^{g_{CO2eq}}/_{MJ}$ for BL and WEU respectively. Figure 5.13 combines these results with the estimates for other common oil sources.

The large uncertainty of ILUC estimates does not allow for solid and precise conclusions about any feedstock in particular; nonetheless, starting from the consolidated observation for which cereals have a lower ILUC effect than oilseeds and at the light of these results, it is possible to conclude that cottonseed-oil RD would lead to consistently less ILUC emissions than the common first-generation feedstocks, such as palm and rapeseed. A striking observation that arises from figure 5.13 is that most oilseed feedstocks, once ILUC effects are included, surpass fossil diesel in terms of GWI. This fact supports the vision of the most recent EU biofuel policies, which puts a cap on such feedstocks. Interestingly, the only two feedstocks that do not overcome such threshold are jatropha, grown on marginal land and thus without any ILUC consequences, and cottonseed oil under the WEU feed supply scheme.

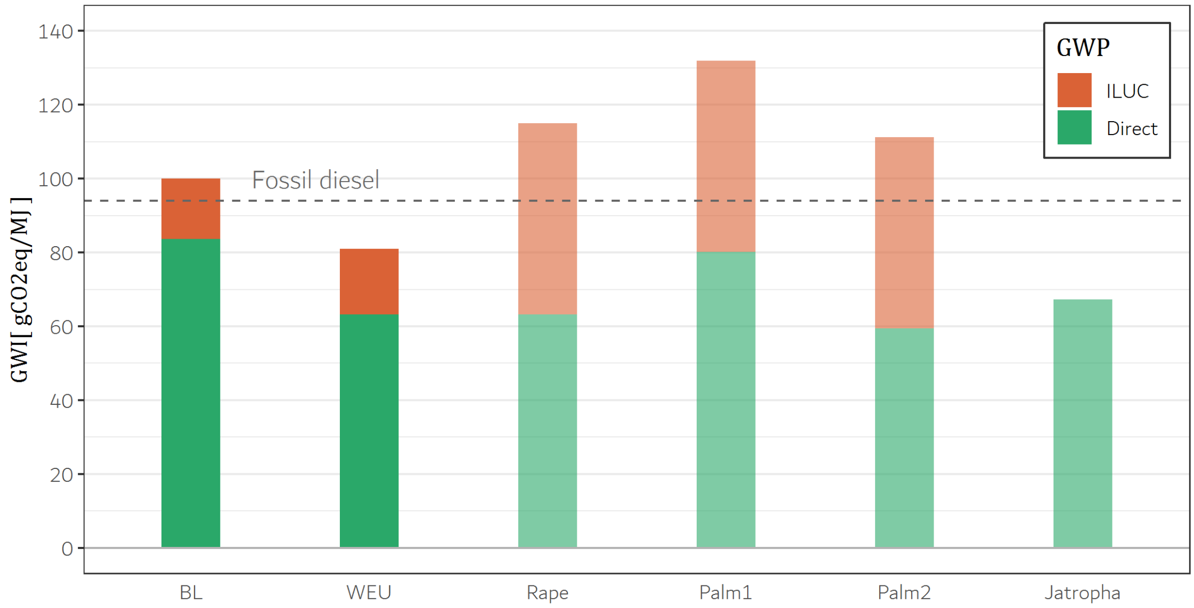


Figure 5.13: Comparison of GWI resulting from the addition of ILUC effects to direct life-cycle emissions (Palm1=palm without CH_4 capture; Palm2=palm with CH_4 capture).

5.3 Conclusions

The present study, which deals with a life-cycle assessment of cottonseed-oil RD production in the specific context of European biofuels regulations, allows to trace conclusions under two distinct viewpoints: a first one, related to process-specific results, and a second one, of wider scope, related primarily to EU regulations and their implications.

The first and foremost conclusion is that cottonseed-oil RD is likely to be compliant with EU biofuels regulations, meaning that its use can effectively concur to the attainment of national and EU-wide renewable target in the transport sector. In the present case study, its GHG emissions reductions exceed 80%, thus well below the required minimum reduction of 50-65%. The scenario analysis seems to indicate that this result is solid (±5% GWI variation) and could be reasonably extended to other contexts, regardless of the previous use of the seed. The main cause for such a life-cycle performance is the regulatory definition of cottonseed as agricultural residue.

Scenarios using ISO guidelines instead of EU rules for developing the LCA lead to a broader set of conclusions. First of all, they indicate that this fuel would reasonably place itself at a GHG emission intensity lower than fossil diesel, although the amount of this reduction would depend to a great extent on the specific cattle feed supplements under consideration. While a certain feed may place the biofuel GWI at an appreciably higher level than other common feedstocks, another supplement may place its GHG intensity in the range of 60 $^{g}{gCO2eq}/_{MJ}$ or lower, in line and even better performing than other common feedstocks. All scenarios also stress the importance of short supply chains: transport of feed supplements represents in fact the dominant cause for transport-related GHG emissions; the case study indicates a potential GWI reduction of around 10 $^{g_{CO2eq}}/_{MJ}$ if grain farm and dairy farm were in direct proximity to each other.

Natural and technical factors - most notably seed oil content, oil extraction yield and reactor selectivity - can have an appreciable influence the results as well: seed oil content is especially relevant, with a registered GWI variation of around $\pm 10-12\%$ within the considered range; this property can be hardly optimised and must be assessed on a case-by-case basis.

An additional, process-specific conclusion is that the inclusion of indirect land use change effects in the life-cycle GHG emissions accounting seems to favour cottonseed-oil-based RD over competing feedstocks. This conclusion stems primarily from the fact that cereals prompt less ILUC emissions than oilseeds and oil palm, a fact on which ILUC studies tend to agree.

From the study, it finally emerges that the methodology used in sustainability assessments – here in terms of climate change – influences the outcomes. EU methodology, while providing an unambiguous set of rules for GHG savings assessment, was not able to include an important portion of the impacts, with a substantial distortion of the results. Regulators must be aware of the possible flaws that a rigid methodology may bring with.

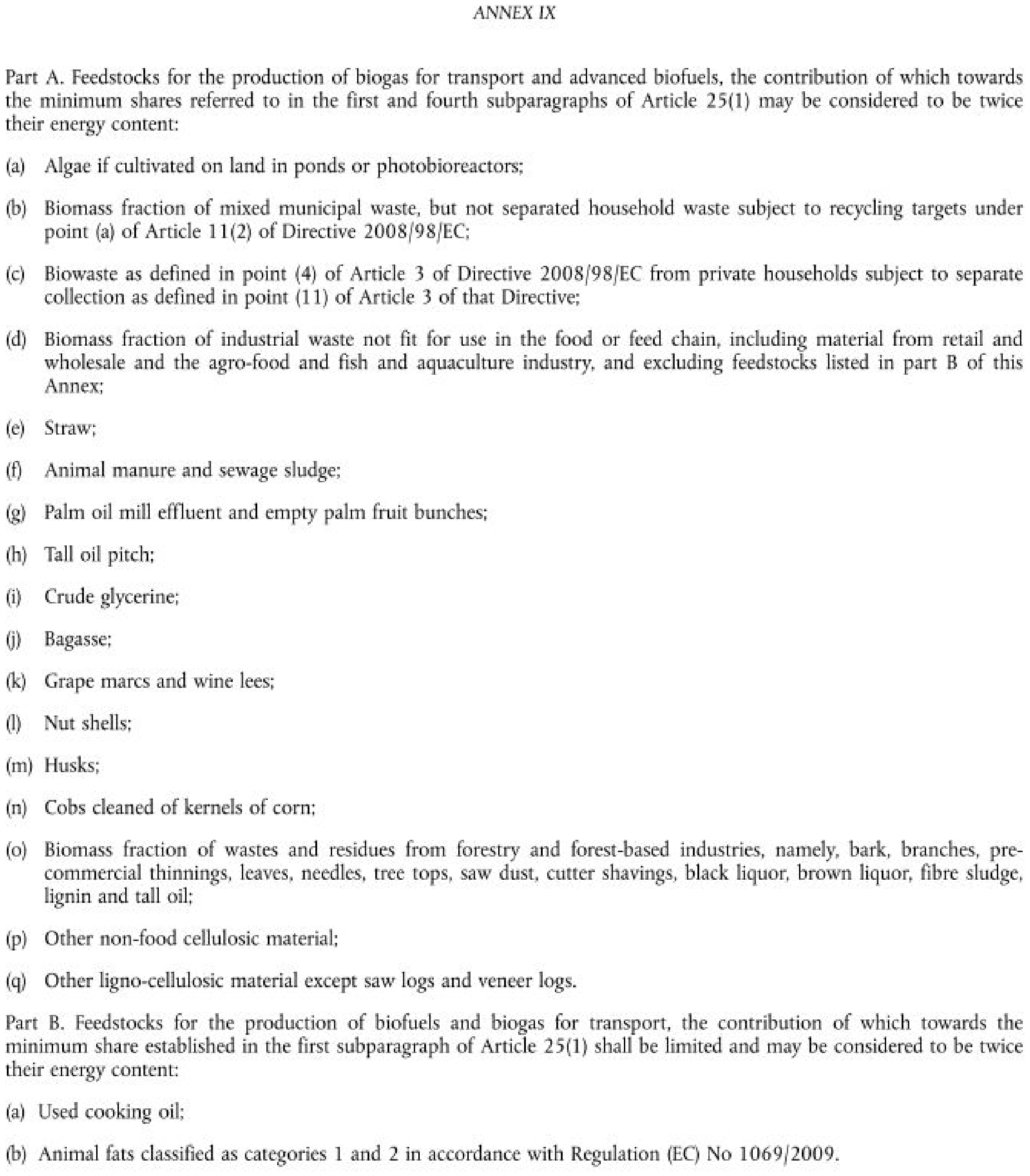
Starting from the latest EU regulations, the study highlights how these may set an especially unfavourable situation for the HVO industry in the near future. The drivers thereof are the difficulty of finding alternatives to palm oil, the limited potential of used cooking oils and fats, and the lack of a regulatory multiplier for oilseeds grown on marginal lands. In this respect, cottonseed may contribute, in the medium term, to fill the gap caused by the phase-out of palm oil. In this respect, Africa appears to be a promising region with yet much unexploited potential of seed being wasted. As presented in section 3.1.3, together with fuel use there are many other possible upgrades for cottonseed and its oil. Given the priorities of many countries in Africa, food and feed industry should be the preferred field of action. Seed toxicity when not further processed, together with the relative simplicity of the supply chain leading to export of oil for biofuels, may advocate in favour of the use as fuel in the medium term; in a second moment, it would be straightforward to upgrade the biofuels pathway to the local food industry, for example, with the addition of a refining process in the country of origin. An adaptation of the LCA model has been tested, in this respect, with the aim of assessing the GHG performance of simple press compared to the more technologically-advanced solvent extraction; in this case, data for a cottonseed crushing facility were available: the plant, located in rural Brazil, utilises firewood as heat source and electricity from the national grid, conditions that could fit many rural contexts also in Africa, like the ones discussed in section 3.1.3. For the purpose of this preliminary analysis, wood-related CO_2 emissions are excluded from the GWI, as if wood is sourced in a renewable way, and not at rhythms that do not match nature's ability to restore the biomass; this choice, in principle, should depend on the specific boundary conditions. The oil yield, in this plant, amounts to 56%; the dataset does not specify the moisture content of inputs and outputs, thus baseline values are kept unchanged. The immediate consequence of this substitution is an increase in meal caloric content: as a consequence, this scenario features lower feed supplement needs per unit of seed, but higher seed needs per unit of fuel. In terms of utility consumption, this process is less impacting, per unit fuel, than the hexane extraction plant featured in all other scenarios. These two phenomena exert opposite effects on system performance, and lead to a higher GWI of RD: this GWI increase is around +25% for the BL supply scheme, provoked almost exclusively by the augmented feed needs. Such increase stresses the importance of a local, carbon-efficient feed supply, especially in rural contexts; here, a case-specific analysis of feed supplements should be carried out, in particular whenever the access to the global market is limited and reliance on local resources is prevalent.

Limitations to the current research, which may be addressed in future work, stem primarily from agricultural data. This research relies in fact on a limited number of certified data, hardly ever accompanied by a statistical distribution. Furthermore, with respect to the specific case study, inventory data for Ukraine, the main exporter of cereals to Israel, are completely missing. Furthermore, the study does not analyse the effects of different agricultural practices such as reduced tillage or cover crops. Direct measurements of N_2O emissions for feed supplements are not available, so that it was necessary to rely on standard emission factors; these do not account for different emissions intensity from different fertilisers. With regards to cottonseed processing, having reliable specific data for an extraction plant processing cottonseed would benefit the precision of the results. Finally, with regards to biorefining, a more punctual modelling would feature a cogeneration unit instead of distinct heat and electricity supplies, but this could not be modelled due to lack of industrial data.

Further research on the topic of cottonseed-oil may focus on specific contexts, especially rural and from developing countries, to assess the sustainability of this biofuel supply chain and to verify its applicability. Other interesting outcomes may come from comparing a large sample of feed supplements in its impact on the fuel GWI, depending on their different origins and agricultural practices. Studies covering impact dimensions other than climate change could give additional evidence on the effective sustainability of this biofuel.

Appendices

Annex IX of EU Recast of the Renewable Energy Directive (RED II).



ANNEX IX

Part A. Feedstocks for the production of biogas for transport and advanced biofuels, the contribution of which towards the minimum shares referred to in the first and fourth subparagraphs of Article 25(1) may be considered to be twice their energy content:

(a) Algae if cultivated on land in ponds or photobioreactors;

(b) Biomass fraction of mixed municipal waste, but not separated household waste subject to recycling targets under point (a) of Article 11(2) of Directive 2008/98/EC;

(c) Biowaste as defined in point (4) of Article 3 of Directive 2008/98/EC from private households subject to separate collection as defined in point (11) of Article 3 of that Directive;

(d) Biomass fraction of industrial waste not fit for use in the food or feed chain, including material from retail and wholesale and the agro-food and fish and aquaculture industry, and excluding feedstocks listed in part B of this Annex;

(e) Straw;

(f) Animal manure and sewage sludge;

(g) Palm oil mill effluent and empty palm fruit bunches;

(h) Tall oil pitch;

(i) Crude glycerine;

(j) Bagasse;

(k) Grape marcs and wine lees;

(l) Nut shells;

(m) Husks;

(n) Cobs cleaned of kernels of corn;

(o) Biomass fraction of wastes and residues from forestry and forest-based industries, namely, bark, branches, pre-commercial thinnings, leaves, needles, tree tops, saw dust, cutter shavings, black liquor, brown liquor, fibre sludge, lignin and tall oil;

(p) Other non-food cellulosic material;

(q) Other ligno-cellulosic material except saw logs and veneer logs.

Part B. Feedstocks for the production of biofuels and biogas for transport, the contribution of which towards the minimum share established in the first subparagraph of Article 25(1) shall be limited and may be considered to be twice their energy content:

(a) Used cooking oil;

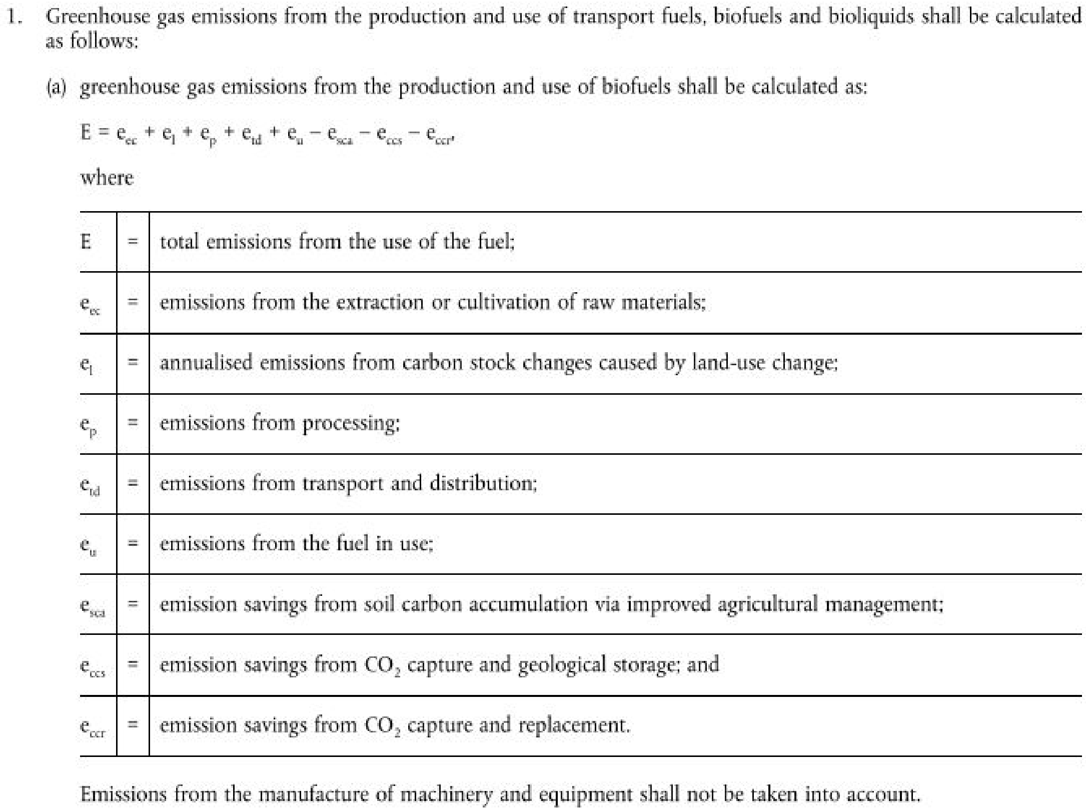
(b) Animal fats classified as categories 1 and 2 in accordance with Regulation (EC) No 1069/2009.

List of multipliers for the attainment of blending targets by EU Member States, in compliance with RED II.

Table B.1: Energy multipliers introduced by RED II for the compliance with the target share of RES in transport final gross energy consumption. [7]

Regulatory element	**Multiplier**
Advanced biofuels (Annex IX, part A)	2
Biofuels from waste oils and fats (Annex IX, part B)	2
Non-food biofuels for aviation	1.2
Non-food biofuels for maritime sector	1.2
RES electricity supplied to rail transport	1.5
RES electricity supplied to road transport	4

Calculation of the emission factor of biofuels according to RED II, annex V



1. Greenhouse gas emissions from the production and use of transport fuels, biofuels and bioliquids shall be calculated as follows:

   (a) greenhouse gas emissions from the production and use of biofuels shall be calculated as:

   $E = e_{ec} + e_l + e_p + e_{td} + e_u - e_{sca} - e_{ccs} - e_{ccr}$,

   where

   E = total emissions from the use of the fuel;

   e_{ec} = emissions from the extraction or cultivation of raw materials;

   e_l = annualised emissions from carbon stock changes caused by land-use change;

   e_p = emissions from processing;

   e_{td} = emissions from transport and distribution;

   e_u = emissions from the fuel in use;

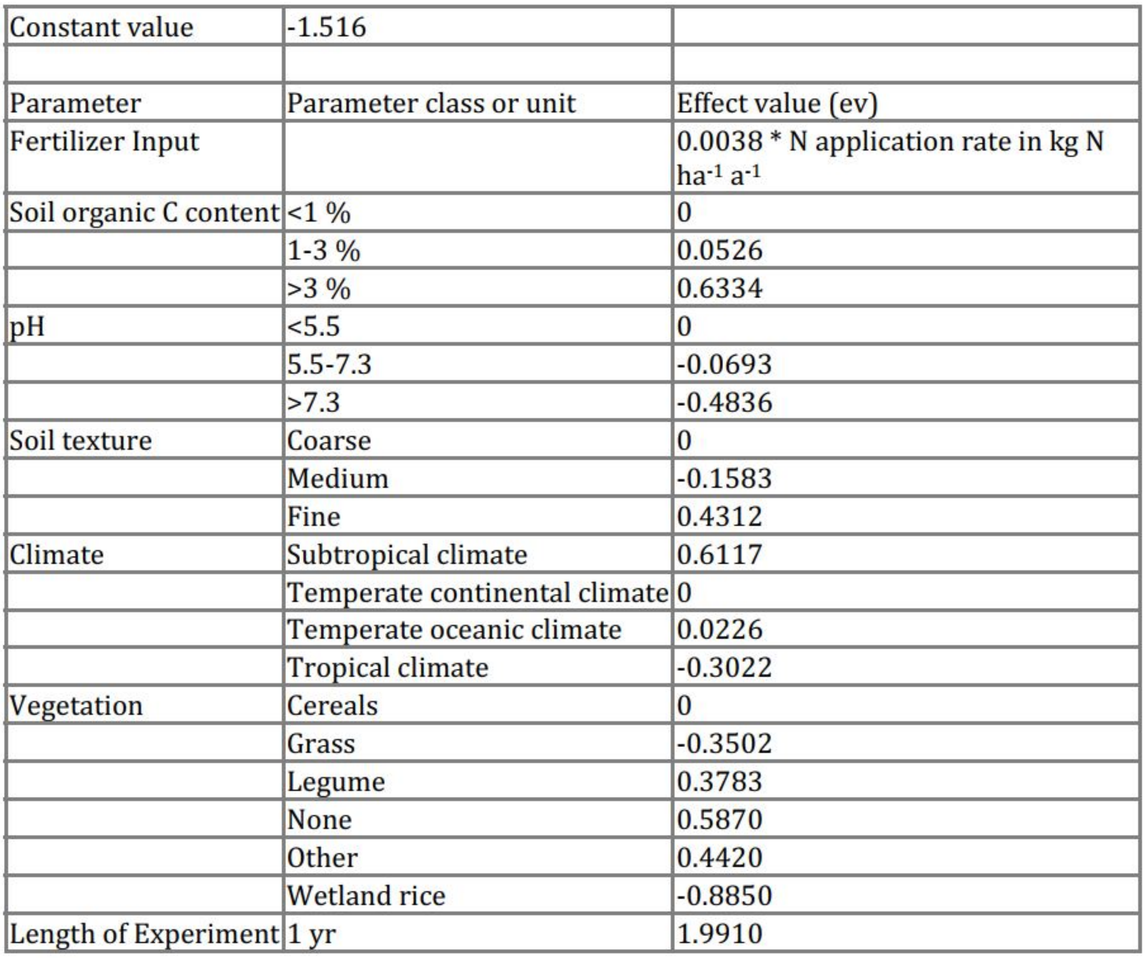
   e_{sca} = emission savings from soil carbon accumulation via improved agricultural management;

   e_{ccs} = emission savings from CO_2 capture and geological storage; and

   e_{ccr} = emission savings from CO_2 capture and replacement.

   Emissions from the manufacture of machinery and equipment shall not be taken into account.

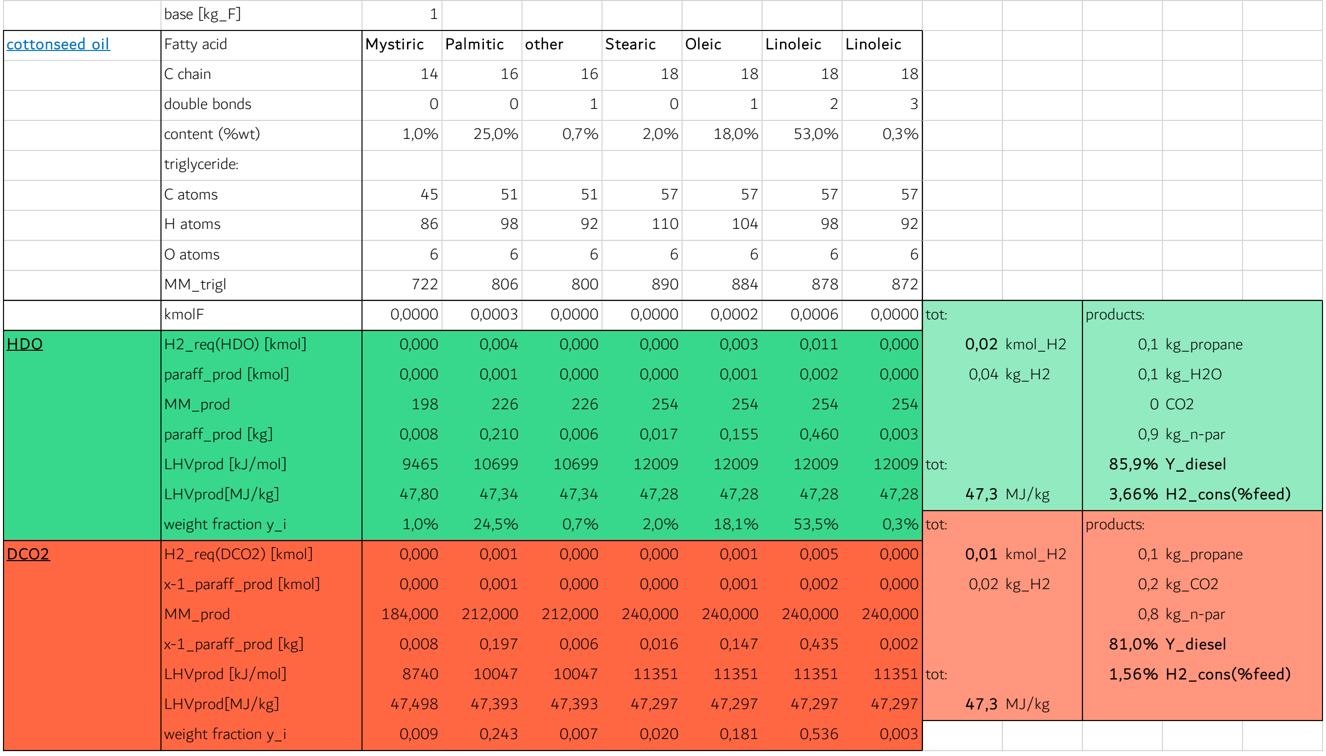
Complete summary of effect values of the Stehfest and Bouwman statistical model



Constant value	-1.516	
Parameter	Parameter class or unit	Effect value (ev)
Fertilizer Input		0.0038 * N application rate in kg N ha^{-1} a^{-1}
Soil organic C content	<1 %	0
	1-3 %	0.0526
	>3 %	0.6334
pH	<5.5	0
	5.5-7.3	-0.0693
	>7.3	-0.4836
Soil texture	Coarse	0
	Medium	-0.1583
	Fine	0.4312
Climate	Subtropical climate	0.6117
	Temperate continental climate	0
	Temperate oceanic climate	0.0226
	Tropical climate	-0.3022
Vegetation	Cereals	0
	Grass	-0.3502
	Legume	0.3783
	None	0.5870
	Other	0.4420
	Wetland rice	-0.8850
Length of Experiment	1 yr	1.9910

Figure D.1: Summary of effect values used to calculate tier 2 direct nitrous oxide emissions for Danish barley in BDK feed supplement scheme.

HDO and DCO2 reactions: feed and products under theoretical conditions



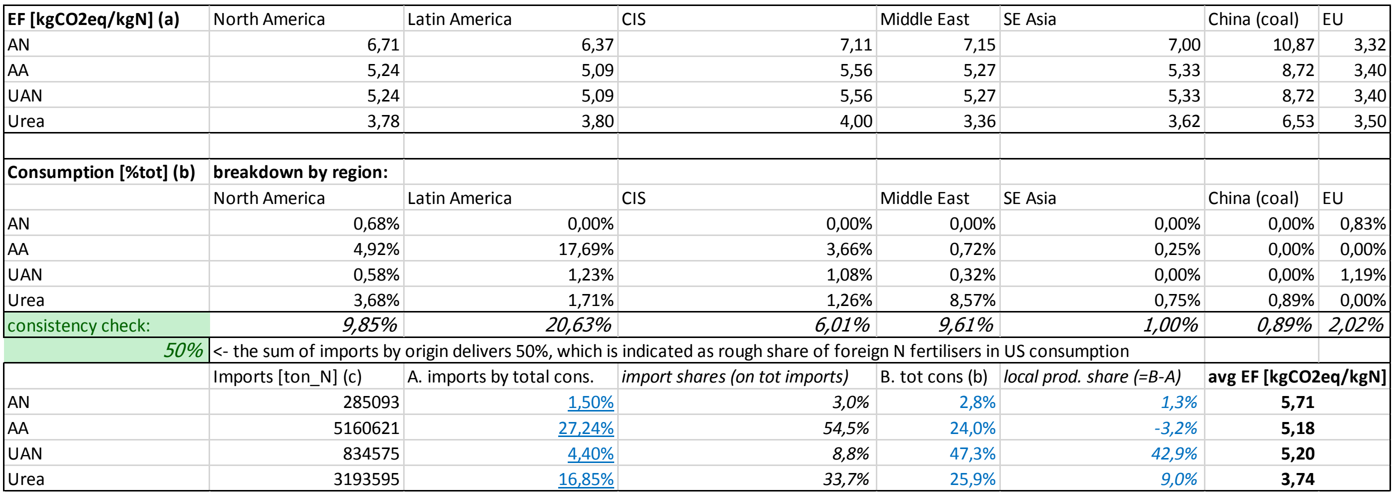
	base [kg_F]	1										
cottonseed oil	Fatty acid	Mystiric	Palmitic	other	Stearic	Oleic	Linoleic	Linoleic				
	C chain	14	16	16	18	18	18	18				
	double bonds	0	0	1	0	1	2	3				
	content (%wt)	1,0%	25,0%	0,7%	2,0%	18,0%	53,0%	0,3%				
	triglyceride:											
	C atoms	45	51	51	57	57	57	57				
	H atoms	86	98	92	110	104	98	92				
	O atoms	6	6	6	6	6	6	6				
	MM_trigl	722	806	800	890	884	878	872				
	kmolF	0,0000	0,0003	0,0000	0,0000	0,0002	0,0006	0,0000	tot:		products:	
HDO	H2_req(HDO) [kmol]	0,000	0,004	0,000	0,000	0,003	0,011	0,000	**0,02**	kmol_H2	0,1	kg_propane
	paraff_prod [kmol]	0,000	0,001	0,000	0,000	0,001	0,002	0,000	0,04	kg_H2	0,1	kg_H2O
	MM_prod	198	226	226	254	254	254	254			0	CO2
	paraff_prod [kg]	0,008	0,210	0,006	0,017	0,155	0,460	0,003			0,9	kg_n-par
	LHVprod [kJ/mol]	9465	10699	10699	12009	12009	12009	12009	tot:		**85,9%**	**Y_diesel**
	LHVprod[MJ/kg]	47,80	47,34	47,34	47,28	47,28	47,28	47,28	**47,3**	MJ/kg	**3,66%**	**H2_cons(%feed)**
	weight fraction y_i	1,0%	24,5%	0,7%	2,0%	18,1%	53,5%	0,3%	tot:		products:	
DCO2	H2_req(DCO2) [kmol]	0,000	0,001	0,000	0,000	0,001	0,005	0,000	**0,01**	kmol_H2	0,1	kg_propane
	x-1_paraff_prod [kmol]	0,000	0,001	0,000	0,000	0,001	0,002	0,000	0,02	kg_H2	0,2	kg_CO2
	MM_prod	184,000	212,000	212,000	240,000	240,000	240,000	240,000			0,8	kg_n-par
	x-1_paraff_prod [kg]	0,008	0,197	0,006	0,016	0,147	0,435	0,002			**81,0%**	**Y_diesel**
	LHVprod [kJ/mol]	8740	10047	10047	11351	11351	11351	11351	tot:		**1,56%**	**H2_cons(%feed)**
	LHVprod[MJ/kg]	47,498	47,393	47,393	47,297	47,297	47,297	47,297	**47,3**	MJ/kg		
	weight fraction y_i	0,009	0,243	0,007	0,020	0,181	0,536	0,003				

Complete inventory for grain production.

	JRC	(Spatari,2020)	(DCA,2016)	NREL	JRC	NREL	NREL	(Kim,2009)
Grain	B	B	B	W	W	M	M	M
Location	EU	US	DK	US	EU	Great Plains	Midwest	Corn Belt
Yield [kg,dry]	3933	5014	5852	2901	5015	8155	8942	7166
N fert. [kgN]	2,44E-02	2,09E-02	1,86E-02	2,56E-02	2,28E-02	1,76E-02	1,75E-02	2,03E-02
P fert. [kgP2O5]	6,10E-03	5,40E-02	3,59E-03	1,22E-02	4,69E-03	1,63E-02	9,97E-03	8,93E-03
K fert. [kg K2O]	6,10E-03	2,51E-02	1,06E-02	1,23E-02	3,67E-03	1,23E-02	1,29E-03	1,17E-02
Liq. Fuels [MJ]	8,27E-01	1,92E-01	6,86E-01	1,42E+00	6,56E-01	3,43E-01	5,50E-01	3,88E-01
Nat. Gas [m3]	8,16E-05	8,16E-05	8,16E-05	3,45E-07	8,16E-05	8,68E-04	1,18E-02	4,86E-03
Lime [kg]	6,17E-02	1,48E-01	0	0	4,36E-02	1,72E-03	9,53E-04	3,21E-03
Electricity [MJ]	0	0	0	2,83E-02	0	1,12E-01	1,68E-02	1,07E-01
Manure [kgN]	0	0	0	0	0	5,52E-04	1,80E-03	0

Figure F.1: Inputs refer to one kg of grain, dry matter, at farm gate. The cut-off criteria have been uniformed to exclude pesticides, herbicides and seedling material.

Reconstruction of the supply of N fertilisers to the US, by type and geographical origin



EF [kgCO2eq/kgN] (a)	North America	Latin America	CIS	Middle East	SE Asia	China (coal)	EU
AN	6,71	6,37	7,11	7,15	7,00	10,87	3,32
AA	5,24	5,09	5,56	5,27	5,33	8,72	3,40
UAN	5,24	5,09	5,56	5,27	5,33	8,72	3,40
Urea	3,78	3,80	4,00	3,36	3,62	6,53	3,50
Consumption [%tot] (b)	**breakdown by region:**						
	North America	Latin America	CIS	Middle East	SE Asia	China (coal)	EU
AN	0,68%	0,00%	0,00%	0,00%	0,00%	0,00%	0,83%
AA	4,92%	17,69%	3,66%	0,72%	0,25%	0,00%	0,00%
UAN	0,58%	1,23%	1,08%	0,32%	0,00%	0,00%	1,19%
Urea	3,68%	1,71%	1,26%	8,57%	0,75%	0,89%	0,00%
consistency check:	*9,85%*	*20,63%*	*6,01%*	*9,61%*	*1,00%*	*0,89%*	*2,02%*
50%	<- the sum of imports by origin delivers 50%, which is indicated as rough share of foreign N fertilisers in US consumption						
	Imports [ton_N] (c)	A. imports by total cons.	*import shares (on tot imports)*	B. tot cons (b)	*local prod. share (=B-A)*	**avg EF [kgCO2eq/kgN]**	
AN	285093	1,50%	*3,0%*	2,8%	*1,3%*	**5,71**	
AA	5160621	27,24%	*54,5%*	24,0%	*-3,2%*	**5,18**	
UAN	834575	4,40%	*8,8%*	47,3%	*42,9%*	**5,20**	
Urea	3193595	16,85%	*33,7%*	25,9%	*9,0%*	**3,74**	

Figure G.1: (a) taken from [30]; (b) taken from [31]; (c) taken from [32].

List of considered transportation processes by mode and distance

Table H.1: Transport processes of fertilisers have been omitted in this list for brevity and relative low relevance. All distances are inclusive of return trip.

Item	**Departure**	**Destination**	**Distance**	**Mode**
Cottonseed	Gin (Migdal Ha'Emek)	Extraction plant	100 km	Truck, 14-20t
Cottonseed meal	Extraction plant	Dairy farm	100 km	Truck, 12-14t
Cottonseed oil	Extraction plant	Port of Haifa	70 km	Truck, 14-20t
Cottonseed oil	Port of Haifa	Port of Gela	3915 km	Container ship
Grains(US)	Farm	Regional storage (Corn Belt)	150 km	Truck, 20-26t
Grains(US)	Regional storage (Corn Belt)	Port of New Jersey	2500 km	Cargo train
Grains(US)	Port of New Jersey	Port of Haifa	19280 km	Container ship
Grains(EU)	Farm	Regional storage	150 km	Truck, 20-26t
Grains(EU)	Regional storage	Port of Constantza	3000 km	Cargo train
Grains(EU)	Port of Constantza	Port of Haifa	3750 km	Container ship
Grains	Port of Haifa	Regional storage	150 km	Truck, 14-20t
Grains	Regional storage	Dairy farm	100 km	Truck, 12-14t

Glossary

AN	Ammonium Nitrate
AP	Available Proteins
AtJ	Alcohol-to-Jet-fuel
BDK	Danish Barley Scenario
BL	Baseline
DCO2	decarboxylation
DCO	decarbonylation
DSHC	Direct Sugars-to-Hydrocarbons
ECCP	European Climate Change Package
EC	European Commission
ETS	Emission Trading Scheme
EUC	EU Compliance Scenario
FAME	fatty acid methyl ester

FFA Free Fatty Acid

GHG Greenhouse Gas

GIS Geographic Information System

GNOC Global crop and site specific Nitrous Oxide emission Calculator

GWI Global Warming Intensity

HDO hydrodeoxygenation

HDS Hydrodesulphurisation

HI High-CO2-Equivalent

HTL Hydrothermal Liquefaction

HVO Hydrotreated Vegetable Oil

ICAO International Civil Aviation Organisation

ICB Israel Cotton Board

ICE Internal Combustion Engine

IEA International Energy Agency

ILUC Indirect Land Use Change

IMO International Maritime Organisation

IPCC Intergovernmental Panel on Climate Change

IS International Standard

ISO International Organization for Standardization

JRC EU Joint Research Center

LCA Life Cycle Assessment

LCIA Life Cycle Impact Assessment

LCI Life Cycle Inventory

LDV Light Duty Vehicle

LNG Liquefied Natural Gas

LO Low-CO2-Equivalent

MS Member States

NEg Net Energy for Growth

NEm Net Energy for Maintenance

PFAD Palm Fatty Acid Distillate

PtL Power-to-Liquid

RD Renewable Diesel

RED II Renewable Energy Directive II

RED Renewable Energy Directive

RES Renewable Energy Sources

SECA Sulphur Emission Control Area

SVO Straight Vegetable Oil

UAN Urea Ammonium Nitrate

UCOAF Used Cooking Oils and Animal Fats

UCO Used Cooking Oils

ULSD Ultra-Low Sulphur Diesel

UNCTAD United Nations Conference on Trade and Development

USDA United States Department of Agriculture

WEU EU Wheat Scenario

WTO World Trade Organisation

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