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Identification of 'Carbon Hot-Spots' and Quantification 1 of GHG Intensities in the Biodiesel Supply Chain using Hybrid LCA and Structural Path Analysis

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Identification of ‘Carbon Hot-Spots’ and Quantification of GHG Intensities in the Biodiesel Supply Chain using Hybrid LCA and Structural Path Analysis

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Abstract

It is expected that biodiesel production in the EU will remain the dominant contributor as part of a 10% minimum binding target for biofuel in transportation fuel by 2020 within the 20% renewable energy target in the overall EU energy mix. Life cycle assessments (LCA) of biodiesel to evaluate its environmental impacts have, however, remained questionable, mainly because of the adoption of a traditional process analysis approach resulting in system boundary truncation and because of issues regarding the impacts of land use change and N_2O emissions from fertiliser application. In this study, a hybrid LCA methodology is used to evaluate the life cycle CO_2 equivalent emissions of rape methyl ester (RME) biodiesel. The methodology uses input-output analysis to estimate upstream indirect emissions in order to complement traditional process LCA in a hybrid framework. It was estimated that traditional LCA accounted for 2.7 kg CO_2-eq per kg of RME or 36.6% of total life cycle emissions of the RME supply chin. Further to the inclusion of upstream indirect impacts in the LCA system (which accounted for 23% of the total life cycle emissions), emissions due to direct land use change (6%) and indirect land use change (16.5%) and N_2O emissions from fertiliser

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applications (17.9%) were also calculated. Structural path analysis is used to decompose upstream indirect emissions paths of the biodiesel supply chain in order to identify, quantify and rank high carbon emissions paths or 'hot-spots' in the biodiesel supply chain. It was shown, for instance, that inputs from the 'Other Chemical Products' sector (identified as phosphoric acid, H_3PO_4) into the biodiesel production process represented the highest carbon emission path (or hot-spot) with 5.35% of total upstream indirect emissions of the RME biodiesel supply chain.

1. Introduction

There has been a growing interest in the use of biofuels as a sustainable replacement for fossil fuels over recent years. This has led to many countries, including the UK and the wider EU community, formulating policies that set out long-term strategies to promote biofuel production and use driven mainly by policy goals such as: reducing greenhouse gas emissions through the decarbonisation of transport fuels, diversifying fuel supply sources and developing long-term replacements for fossil oil. The EU has a long-term vision for biofuels, proposing that by 2030 and beyond, clean and CO_2-efficient biofuels would make up 25% of the EU's transport fuel needs [1]. Refer to Figure S1 in the Supporting Information (SI) on the web for an illustration of the transition plan of past EU policies affecting biofuels and the timescale for future commitments.

Biodiesel is Europe's dominant renewable fuel [2] with rapeseed accounting for about 80% of primary feedstock for biodiesel processing and about 75% share of total oilseed production of EU-27 in 2009-10 [3]. Production of biodiesel on an industrial scale began in 1992 about

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five years before the EU's Energy White Paper 'Energy for the Future', driven mainly by positive signals in terms of support from member states and the EU Commission. Figure S2 under SI on the web shows the trend in the growth of biodiesel in the EU since 1992.

Despite the potential growth and benefits of biofuels, many research findings have raised arguments against them in a global context. The Food and Agricultural Organisation [4] acknowledges that, although biofuels under certain conditions help reduce greenhouse gas emissions (GHG), the global effects of an expansion of biofuel production will depend crucially on where and how feedstocks are produced. It is therefore anticipated that the more sustainable second-generation biofuels produced from non-food crops and residues can provide the best opportunity for the commercial viability and development of the sector from 2014 onwards [5]. Lifecycle assessment (LCA) of second-generation bioethanol produced from surplus forest-bioenergy resources in Norway for example was estimated to potentially save 6-8% of Norway's global warming GHG emissions associated with road transportation [6]. The accelerating use of biomass including, cereals such as wheat, maize, sugar and oilseed for biofuel and power generation has come about because of positive government directives and political decisions [7]. However, the exact impact on the resource base and the environment due to the demand for biofuels is unknown.

Many authors have therefore undertaken studies to evaluate the environmental impacts of biofuel production [8-10]. These studies have mainly used traditional LCA methods based on ISO 14040 and mostly involved comparative studies with traditional fossil fuel production [11]. Traditional LCA of biofuel production involves setting a system boundary for the

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biofuel supply chain and using process analysis data to estimate the carbon impacts of selected supply chains within the system boundary.

It is, however, well recognised that because of difficulties in collecting process-specific data in LCA and the infinite number of possible supply chain paths, the use of hybrid LCA provides a more comprehensive framework for the evaluation of environmental impacts of upstream production [12-15]. A hybrid LCA combines the specificity of process analysis with the extended system boundary of input-output (IO) analysis. Hybrid LCA has had many applications. Lenzen and Wachsmann [16] and Crawford [17] demonstrated the use of a hybrid LCA technique in the assessment of the energy content of wind turbines in order to achieve system completeness. A limited number of studies using hybrid LCA have been undertaken on biofuels. Bright et al. [18] undertook an environmental assessment of wood-based biofuel to estimate the cumulative global warming mitigation under different scenarios in Norway. The hybrid LCA in this study consisted of a two-region (Norway and the European Union) IO model and process analysis inventory for the biofuel options.

In this paper the life cycle GHG emissions of a typical biodiesel supply chain are calculated using hybrid LCA, incorporating process-specific data of rape methyl ester (RME) production and inputs from higher upstream processes such as chemical inputs, mining, transportation, banking, equipment, etc, based on input-output analysis. Direct and indirect emissions in the biofuel supply chain are determined, including direct and indirect land use change and N_2O emissions from fertiliser application. Furthermore, structural path analysis (a decomposition technique used in economic and ecological systems analysis) is applied to identify, quantify and rank high carbon emission paths – or 'carbon hot-spots' – in the supply chain. Some studies have demonstrated the use of SPA as a decomposition technique of environmental impacts in a LCA context [19-20]. To the best of the authors' knowledge, this is the first time that SPA has been used for an analysis of the biodiesel supply chain. This detailed analysis is aimed at helping to tailor and prioritise mitigation efforts through the use of biofuels.

2. Material and Methods

'Integrated hybrid LCA' as defined by Suh and Huppes [21] is applied in this study. This form of hybrid LCA combines a process matrix and an IO matrix in a consistent mathematical framework [22]. Whereas the process component systematically computes physical inputs and outputs of each production step within the system boundary, the input-output component completes the analysis by enumerating upstream indirect inputs from outside the process system boundary.

For an integrated hybrid assessment of biofuel supply chains, the process matrix is linked to the input-output matrix using the operational expenditure of biofuel production to account for upstream inputs. As shown by Suh and Huppes [21], the general relationship for the integrated hybrid model is given in matrix notation by:

$$\widetilde{\mathbf{P}}_{\mathbf{hybrid}} = \begin{bmatrix} \mathbf{E_p} & \mathbf{0} \\ \mathbf{0} & \mathbf{E_{i-o}} \end{bmatrix} \begin{bmatrix} \mathbf{A_p} & -\mathbf{D} \\ -\mathbf{U} & (\mathbf{I} - \mathbf{A_{i-o}}) \end{bmatrix}^{-1} \begin{bmatrix} \mathbf{y} \\ \mathbf{0} \end{bmatrix} \qquad \text{(Equation 1)}$$

Where:

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$\widetilde{P}_{hybrid}$ = Total (direct and indirect) environmental impact (e.g. CO_2-eq emissions) associated with one unit of final demand y for the product (here biodiesel).

A_p = square matrix representation of process inventory, (dimension: $s \times s$)

A_{i-o} = IO technology coefficient matrix (dimension: $m \times m$)

$\mathbf{I}$ = identity matrix (dimension: $m \times m$)

$\mathbf{U}$ = matrix representation of upstream cut-offs to the process system (dimension: $m \times s$)

$\mathbf{D}$ = matrix of downstream cut-offs to the process system (dimension: $s \times m$)

$E_{\mathbf{p}}$ = process inventory environmental extension matrix. CO_2-eq emissions are diagonalised (dimension: $m \times s$)

$E_{\mathbf{i-o}}$ = IO environmental extension matrix. CO_2-eq emissions are diagonalised (dimension: $m \times s$)

$\begin{bmatrix}\mathbf{y}\\\mathbf{0}\end{bmatrix}$ = Functional unit column matrix with dimension $(s + m, 1)$ where all entries are $\mathbf{0}$ except $\mathbf{y}$

Matrix A_p describes the product inputs into processes as captured in the unit process exchanges (or process analysis inventory from ecoinvent in this case) and described in Table S1. These processes, together with the sectoral inputs from IO sectors, are used to draw up the biodiesel supply chain map as depicted in Figure S3.

Matrix U, which is assigned a negative sign, represents the higher upstream inputs from the IO system to the process system. Matrix D, also assigned a negative sign, represents the (downstream) use of goods / process inputs from the process to the background economy (IO system). As explained by Suh and Huppes [21], the downstream cut-off matrix represents the link from the process-based (foreground) system to the IO-based (background) system. It can be argued that the downstream cut-off flows in D are often small compared to the – normally much larger – background economy (cf.[23]). The aim of the present paper is to quantify the total emissions of biodiesel production in the status-quo economy of the UK in 2004 when the market share of biodiesel as a percentage of road transport fuel was at a modest 0.09% in 2004 [24]. For the sake of simplification we therefore neglect interactions with the background economy and set values in D are set to zero. We acknowledge, however, that a more general use of biodiesel in the economy would ideally be evaluated by including industries' expenditures on biodiesel in D (e.g. by assuming different market penetrations of biodiesel in a number of scenarios).

The final demand $\tilde{\boldsymbol{y}}$ for biodiesel also represents the functional unit of the LCA system, set to 1kg of RME biodiesel in this study.

In order to achieve a complete LCA system for the biodiesel supply chain, upstream cut-offs from the process-based LCA system were estimated using input-output analysis. For example, to estimate the contributions of an upstream service (for example: administration) for a given process inventory (for example: electricity) already captured in the process matrix, $\mathbf{A_p}$ the following steps were taken. The unit cost of the process under consideration (example: electricity) was obtained [£/kWh]. This was multiplied by the input (in physical terms) of electricity [kWh] obtained from the process matrix. The results, k (that is: [£/kWh]* [kWh]) represents the amount of electricity (in £) needed to produce 1kg of final demand of biodiesel. This amount is then used as a scalar multiplier to the column a_{ij} of the IO technology matrix, A_{i-o} where j corresponds to the Electricity industry. To avoid double counting, all inputs already captured in the process matrix are discounted from the resulting column vector ka_{ij}. The corrected values $ka_{ij}*$ become elements of the upstream input matrix *U*. The administrative expenditure linking the process LCA electricity to the IO table corresponds to ka_{ij} where i corresponds to Administration as a product and j Electricity as an industry. Refer to Spreadsheet S1.

Uncertainty in upstream emissions was estimated by including the maximum/minimum IO upstream cut-offs into the LCA system. To account for the maximum IO upstream cut-offs, all potential sectoral products that are indirect input requirements into biodiesel production are included. Similarly, to account for minimum IO upstream cut-offs, only sectoral products that are highly probable indirect input requirements into biodiesel production supply chain are included. Refer to the supplementary Spreadsheet S1 for inputs into the upstream supply chain for the maximum and minimum case scenarios. Besides its mathematical consistency, integrated hybrid LCA provides a comprehensive framework because all inputs associated with the biodiesel supply chain can be expressed by the combination of process and IO matrices.

2.1 Structural Path Analysis

Taylor's series expansion is applied only to the IO part of Equation (1) because the inputs of the unit process obtained from ecoinvent are clearly known:

$$P'_{IO} = [E] \times (I - A)^{-1} y \;=\; \boldsymbol{E}\mathbf{Iy} + \boldsymbol{EA}^{\mathbf{1}}\mathbf{y} + \boldsymbol{EA}^{\mathbf{2}}\mathbf{y} + \boldsymbol{EA}^{\mathbf{3}}\mathbf{y} + \cdots + \boldsymbol{EA}^{\mathbf{n}}\mathbf{y} \qquad \text{(Equation 2)}$$

EIy', represents the direct GHG emissions emitted (at production level 0) for a given demand and $E\mathrm{A}^{\mathrm{n}}\mathrm{y}$ the indirect GHG emissions emitted for a given final demand at the n^{th} production level. The Taylor series expansion of the Leontief inverse matrix can be further decomposed by unravelling the A matrix (or the IO technology coefficient matrix with elements, a_{ij}) into a series of structural paths at the n^{th}order to systematically identify important supply chains [25]. Summing up across all products i and industries, j, the total environmental impact of a final demand bundle y_i can be decomposed into: (Equation 3)

$$P_{\mathrm{IO}} = \sum_{i=1}^{\mathrm{n}} \sum_{\mathrm{j}=1}^{\mathrm{n}} e_j \left(I_{ji} + a_{ji} + \sum_{k=1}^{n} a_{jk}\, a_{ki} + \sum_{l=1}^{n} \sum_{k=1}^{n} a_{jl}\, a_{lk} a_{ki} + \cdots \right) \mathrm{y_i}$$

where e_j is the emission intensity of industry j and elements a_{nm} represent transaction coefficients between sector n and m. Each multiplied term represents the contribution of an individual supply chain path. In the case of biodiesel in this study, the emission 'carbon hot spots' in the supply chain were to be identified and therefore the combined upstream inputs from matrix U as the demand bundle y_i were used: (Equation 4)

$$y_i = \sum_{j=1}^{n} k a_{ij} *$$

Where k represents the £ equivalent needed by industry j to produce 1kg of biodiesel.

The decomposition of the series expansion can be represented as a tree diagram (Refer to Figure S4) whereby each tier in the tree represents a different production layer and each node gives the contribution to total environmental impacts from the demand, y [26].

Production layer refers to the stage of supply to the main product. Production layer 0 therefore refers to the biodiesel production process. Production layer 1 is the first stage of the upstream supply chain and production layer 2, is the supplier to the first upstream supplier of the biodiesel process. In a SPA disaggregation of a product system, each node represents a contribution to the total environmental impacts from the demand, y.

The maximum number of nodes at a production level is given by:

$$Number\ of\ Nodes = n^{l+1} \qquad \text{(Equation 5)}$$

n = Number of sectors in the economy

l = Production level

The importance of supply chain decomposition in disentangling upstream emission paths in a product system is evident in the fact that upstream environmental impacts are often greater than direct environmental impacts in a supply chain. In a carbon footprint case study of economic sectors in Australia and the US, Huang et al. [27] for example, showed that direct emissions of the majority of sectors are below 20% of the total carbon footprint, and can be as low as 1%. To maximise the potential for biodiesel to achieve real CO_2 emissions reductions compared to fossil fuels, high emissions intensity paths or 'hot-spots' in the supply chain must be identified and possible lower emission alternative processes for the production of biofuels must be found.

2.2 Process Analysis Data

The 2010 ecoinvent database v2.2 was used to compile the process analysis life cycle inventory described as unit process exchanges. This dataset includes production of biodiesel rape methyl ester (RME) from rape oil from esterification plants in the EU. The operation of storage tanks and fuel stations, including the distribution to the final consumer and all necessary transport requirements are included. Emissions arising from evaporation and treatment of effluents (may also refer to the air emissions of the plant) are also included. For the analysis, corresponding CO_2-eq emissions data of unit process exchanges, E , emitted in producing 1kg of RME biodiesel were determined. Biogenic CO_2 was captured in the unit process exchanges obtained from ecoinvent [28]. It was calculated using the principle of carbon balance (input of carbon = output of carbon); that is, the uptake of carbon during plant growth plus all inputs of biogenic carbon with all pre-products minus biogenic carbon emissions should equal the biogenic carbon content of the biofuel or the product after all allocations have been done [28]. The unit process exchanges representing the process analysis data from ecoinvent are presented as Table S1 as part of Supplementary Information on the web.

2.3 Input-Output (IO) Analysis Data

Previously constructed 2004 UK domestic and UK imports supply and use tables disaggregated to 178 sectors were used to derive the input-output data used in the study [29].

Wiedmann et al. [29] describe the construction of a multi-regional input-output (MRIO) model using UK national IO tables and rest-of-world (ROW) tables from the Global Trade Analysis Project (GTAP). A technology coefficients matrix was derived for both the UK domestic and UK imports use table. For the purpose of the present study, the ROW economy is represented as one symmetric table (technical details of this 2-region model have been described in [30].

Columns of UK and ROW industry input requirements are augmented with data for greenhouse gas emissions to derive sectoral emissions intensities (kg CO_2-eq/£) for the environmental matrix, E_{i-o}. A supply chain map illustrating the comprehensive system boundary framework of the biodiesel supply chain adopted in this study is available in Figure S3 under SI on the web.

2.4 Allocation Factors

The production of RME results in multiple product outputs. For example, the processing of oil mill into rape oil also results in the production of rape mill as a by-product. The esterification of vegetable oil into RME also produces glycerine and potassium sulphate. In order to deal with multiple product outputs, LCA studies apply the method of either allocation or system expansion. In the first case, inventory data are allocated to the main product, by-products and waste, respectively, in order to assign material inputs and environmental impact. In system expansion, the boundary is extended to account for the input and output flows of all products. In this study, we use the first option, allocation, as we are specifically interested in the provision of biodiesel.

Allocation factors can be based either on mass flow, energy value or economic revenue of co-products. Economic allocation has been established as a recognised way of systematically executing allocation in LCA [31-33]. The International Standards Organisation [34] also gives this allocation option in Step 3 of its allocation procedure. Hence, in this study, the economic revenue allocation as adopted in ecoinvent [28] was used. To reduce the uncertainty related to economic allocation because of potential fluctuations in the economic values of product and co-products, the environmental burdens are allocated according to the revenue of all process products, based usually on the average prices for three consecutive years. (Refer to Table S2 for allocation details). Allocation factors for other methods related to the production of RME are also presented in ecoinvent [28].

3. Results and Discussions

3.1 Hybrid Life Cycle CO_2-eq of Biodiesel Production

The total emissions of all unit process exchanges representing the process analysis data of the biodiesel production process is 2.7 kg CO_2-eq or 36.6% of the total life cycle emissions. IO upstream indirect emissions (for the base case impact scenario) account for 1.7 kg CO_2-eq of the total life cycle emissions. Upstream emissions include embodied emissions such as those associated with utilities, equipments, chemicals, mining, construction of buildings, maintenance, services such as banking and finance, insurance, research and development, advertising, etc., and accounted for approximately 23% of total emissions. A further breakdown of these emissions is provided in Section 3.3 (Structural Path Analysis of Biodiesel Supply Chain Emissions). Refer also to Table S3 and Figure S6 on the web.

It was also estimated (from the process analysis inventory in ecoinvent) that the esterification of vegetable oil to RME process accounted for 35.5% of the total emissions or 97% of emissions due to the unit process exchanges in the process inventory. The other unit process exchanges: road and train transport, electricity supply, regional distribution of oil, waste management and water treatment from the process analysis inventory in ecoinvent collectively accounted for around 1.1% of the total emissions associated with the RME biodiesel production process.

3.2 Other Impacts

It has generally been argued that greenhouse gas releases from land use change and nitrous oxide (N_2O) emissions from the use of fertilisers can potentially be significant enough to change the environmental profile of biodiesel [35-36].

With N_2O having a global warming potential 298 times that of CO_2 when considered over a 100-year period [37], the use of nitrogen fertilisers has the potential to significantly affect the GHG emissions balance of biodiesel. N_2O is emitted both directly from soils due to the use of nitrogen-based fertilisers and microbial transformations of organic nitrogen (N), and also indirectly with nitrogen losses through volatilization, leaching and runoff of N-compounds that are converted into N_2O off site.

Also, the European Commission Joint Research Centre [38], Searchinger et al. [39] and Fargione et al. [40] have all stated that indirect land-use change could potentially release enough greenhouse gases to negate the savings from conventional biofuels. Land use can be defined as the type of activity being carried out on a unit of land and the change in land use can be either direct or indirect. Direct land-use change occurs when feedstock being cultivated for biofuels production (e.g. rapeseed for biodiesel) displaces a prior land use (e.g. forest), thereby generating possible changes in the carbon stock of that land. Indirect land-use change on the other hand occurs when pressure on agriculture due to the displacement of previous activity or use of the biomass induces land-use changes on other land [11].

3.2.1 Estimation of N_2O Emissions from Fertiliser Application

The Intergovernmental Panel on Climate Change, IPCC [41] estimate that the direct emission factor associated with N_2O emissions is likely to be 1% of the N applied to the soil. Jungbluth et al. [28] also estimated a direct emissions factor of 1.25% of the N-input and an indirect emission factor of 2.5% from the nitrogen that is leached as nitrate. Crutzen et al. [42] however state that global analysis of N_2O emissions had been previously underestimated and shows that N_2O emission factor (direct and indirect) in agro-biofuel production is 3–5% of N applied (that is 0.03-0.05 kg N_2O-N $(kg\ N)^{-1}$. The maximum value of 5% in Crutzen et al. [42] is used to assess the highest impact case scenario in this study. The minimum value of 3% Crutzen et al. [42] is applied to determine the uncertainty range between the maximum and the minimum impact scenario. Refer to Smeets et al. [43] for additional information on the contribution of N_2O to the greenhouse gas balance of first-generation biofuels. The fertilizer input rate assumed in this study (137.4 kg-N ha^{-1} yr^{-1}) was used in the JEC - Joint Research Centre-EUCAR-CONCAWE collaboration on biofuel programme [44]. In the cultivation of 1kg of rape, the following conditions were assumed:

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i. Land Use: Transformation from non-irrigated arable land accounted for 71% of land use at 2.08 m^2 per kg of rape while transformation from pasture and meadow land accounted for 29% of land use at 0.85m^2 per kg of rape [45]

ii. Land Occupation: 11 months per year permanent land-use occupation [46].

iii. It was deduced from ecoinvent [28] that 2.6 kg of rapeseed is required to produce 1kg of RME biodiesel

The relationship used to determine the life cycle CO_2-eq emissions associated with the use of nitrogen fertilizers can be expressed as: Equation 6

$$\text{Life Cycle Emissions due to } N_2O \text{ Use} = e_f y f_i r_s k_n (GWP)_{N_2O} \sum_{i=1}^{j} l_i \, p_i$$

Where:

e_f = N_2O emissions factor [kg.N_2O–N per kg-N]

y = time between planting and harvest of the bioenergy crop [years]

f_i = Fertilizer input rate [kg-N ha^{-1} yr^{-1}]

r_s = ratio of the kg of rapeseed to required to produce 1kg of RME

k_n = Factor to convert from N_2O-N to N_2O [equivalent to $(\frac{44}{28})$]

$(GWP)_{N_2O}$ = Global Warming Potential of N_2O.

l = Area of land occupied by bioenergy (biodiesel) crop [m^2 per kg of rapeseed]

p = ratio of the area of particular land type occupied by bioenergy crop to the total area of different land used (in cases where only one land type is used, p will be 1)

Using a GWP of 298 for N_2O and effective land size of 1.723 m^2 or (1.723×10^{-4} ha) per kg of rape seed, it is estimated from Equation 6 that the N_2O emissions contributed between a high impact scenario of 1.32 kg CO_2-eq per kg of RME and a low impact scenario of 0.80 kg CO_2-eq per kg of RME biodiesel.

3.2.2 Direct and Indirect Land Use Change

The Intergovernmental Panel on Climate Change [47] reports that due to direct land use change, changes in carbon stock per hectare of bioenergy crop cultivation occurs in the following carbon pools for cropland: biomass (above ground biomass and below ground biomass); dead organic matter (dead wood and litter) and soils (soil organic matter). The total change in carbon stock is calculated using Equation 7 below [10].

Equation 7:

$$\Delta \text{ Carbon Stock Soil } \left[\frac{tC}{ha}\right] = \sum_i \text{Carbon Stock Change Factor } \left[\frac{tC}{ha.yr}\right] \times T_i[yr]$$

Where

T_i= time between planting and harvest of the bioenergy crop [years]

Based on data on carbon stock change factors for the carbon pools of cropland from the IPCC Guidelines for National Greenhouse Gas Inventories – Agriculture Forestry and other Land Use [47], direct land use change was estimated to be 0.44 kg CO_2-eq per kg of RME.

Indirect land use change (iLUC) is calculated using the theoretical global average indirect land use change factor [48]. In this study it was assumed that the cultivation of rape seed occurred on 71% arable and 29% pasture and meadow land. A 'maximum risk' or 'maximum iLUC order of magnitude' representing a 75% share of non-zero risk biofuel is assigned an iLUC factor of 15 t CO_2-eq/ha/yr while a 'low risk' or 'low iLUC order of magnitude' representing 25% of all non-zero risk biofuels are subject to theoretical full iLUC factor of 5 t CO_2-eq/ha/yr [48]. The term risk refers to the level of impact due to the conversion of food crop land into bioenergy crop land. A low risk biofuel is therefore assumed to be produced from feedstock cultivated on set-aside or abandoned land. By weighting these iLUC factors according to the ratio of land type and land sizes assumed in the cultivation of rape seed in this study, the iLUC factor used for to the production of 1 kg of RME biodiesel and its co-products is estimated to be 11.8 t CO_2-eq/ha/yr or 1.86 kg CO_2-eq. Taking into account the allocation factors in Table 2, iLUC is calculated to be 1.22 kg CO_2-eq per kg of RME.

Uncertainty in the impact of land-use change refers to the variability of indirect land-use change factors due to the type of land used in the cultivation of feedstock. Based on the assumptions for maximum/minimum iLUC risk referred to above, the uncertainty range for iLUC for producing 1kg of RME biodiesel is estimated to be between of 0.52 to 1.55 kg CO_2-eq.

The emissions associated with all stages of the RME biodiesel production are shown in Figure 1. The total life cycle CO_2-eq emissions for 1kg of RME biodiesel were calculated to be 7.38 kg CO_2-eq or 199 g CO_2-eq/MJ. By accounting for uncertainty in the assessment, it was estimated that the results are in the range 5.03 to 8.44 kg CO_2-eq per kg of RME biodiesel. Refer to Figure S5 for normalized results in energy units.

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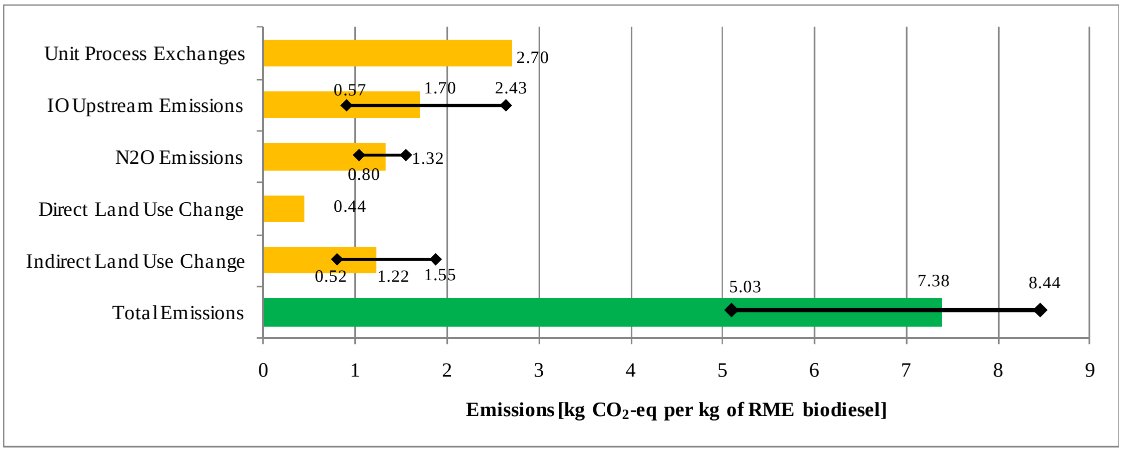
Figure 1: Life Cycle Emissions of RME Production and Supply Chain



3.3 Structural Path Analysis and Hotspots of Biodiesel Supply Chain Emissions.

In Figure S6, the cumulative impacts of sectoral emissions from the higher upstream supply chain paths of biofuel production are presented giving an indication of the relative contribution of each IO sector. These higher upstream supply chain paths represent the IO component of upstream inputs. Seven production layers of RME biodiesel were analysed. It was estimated that the 'Utilities Sector' was the highest sectoral emitter accounting for 172 g CO_2-eq or 44.5% of total upstream emissions. This was followed by the 'Chemical Sector' emitting 90 g CO_2-eq or 23.3% of total upstream emissions. As can be seen from Figure S6, the next four sectoral emissions were 'Transportation and Communication' (37 g CO_2-eq or 9.6%), 'Mining' (21 g CO_2-eq or 5.4%), 'Minerals' (19 g CO_2-eq or 4.8%) and 'Fuels' (14 g CO_2-eq or 3.7%).

Structural path analysis (SPA) is used to show the inter-connections of various products and industries within the biodiesel supply chain and identify, rank and estimate the CO_2-eq of the high emissions intensity paths or 'carbon hot-spots'. 150 of the most important paths of the

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biodiesel supply chain were extracted in the SPA. The cut-off threshold for individual path contributions was set at 0.05% of total impacts in the analysis of the supply chain paths. Detailed results for each of the top 50 paths are shown in Table S3 under SI on the web.

It was found that CO_2-eq emissions impacts on the biodiesel supply chain originate across the entire economy but of the top 150 paths, the majority originate from the sectors 'Other Chemical Products', 'Organic Basic Chemicals', 'Electricity-Coal', 'Distribution and Trade', 'Electricity-Gas' and 'Freight transport by Road'.

The "hottest spot" or the highest carbon intensity path of the biodiesel upstream supply chain was identified as a path order 1: Rest of World (ROW) Sector (102) 'Other Chemical Products' > Biofuel Process with an estimated 20.7 g CO_2-eq or 5.35% of the total emissions. This path describes the emissions chain: 'Other Chemical Products' used as an input in the biodiesel production process.

4. Discussion

The life cycle assessment of the RME biodiesel supply chain estimated the total life cycle emissions of biodiesel production to be 7.38 kg CO_2-eq per kg with an uncertainty range of 5.03 to 8.44 kg CO_2-eq per kg. The uncertainty was a result of variability in indirect land use change, N_2O emissions and IO higher upstream emissions. IO higher upstream emissions accounted for approximately 23% of total CO_2-eq emissions. The use of a hybrid method ensured the integration of process and IO analysis such that higher upstream inputs into sectors such as utilities, transportation, chemicals, mining, services, etc which are normally excluded from traditional life cycle assessments, are taken into account. In contrast, Halleux et al. [8], Hoefnagels et al. [10] and Kim et al. [49] all undertook life cycle assessments of biofuels using traditional LCA but did not account for upstream emissions outside the process system boundary resulting in the truncation of the product system. Given that some past assessments of biofuels have also neglected the impacts of land use change and N_2O emissions, assuming that the impacts of land use change, N_2O emissions and IO upstream emissions cut-offs were truncated from the biofuel product system, as has previously been the case, the total emissions would have been 2.7 kg CO_2-eq per kg. This would have resulted in a 63% underestimation of the total life cycle emissions. Therefore, IO upstream emissions cut-off, N_2O emissions and land-use change represent significant impacts which are determinants that can change the environmental profile of the biodiesel supply chain. It was observed that the lack of process-specific data increases uncertainties in life cycle assessments. The uncertainty estimates for this study were based on data variability in indirect land use change, N_2O emissions from fertiliser application and aggregation of IO data, resulting in the estimation of minimum and maximum carbon impacts of the RME biodiesel supply chain. The estimation of emissions was based on economic allocation between the RME biodiesel supply chain and co-products. This has been recognised as one way of systematically executing allocation in LCA [31-33].

Structural path analysis (SPA) is useful in describing and characterizing carbon hotspots in the supply chain. Specific processes in the RME biodiesel supply chain can be matched to the structural paths in order to identify the hotspots in the supply chain. For example, the first ranked structural path: Other chemical products > Biofuel Process can be identified as the inputs of industrial grade phosphoric acid, H_3PO_4 (85% in water) into the biodiesel production process. Likewise, the second ranked structural path: Organic basic chemicals > Biofuel Process describes the inputs of methanol into the biodiesel production process. As can be seen from the ranked structural paths available in the Table S3, all the paths end as a direct input into the RME diesel production process. The demanding sector is therefore responsible for the emissions caused, but the emissions might occur upstream of that sector. For example, in the 12th ranked structural path: UK- Electricity - Coal> UK- Distribution and Trade in Electricity > Biofuel Process; the biodiesel production process is responsible for emitting 3.28 g CO_2-eq per kg of RME biodiesel although it occurs upstream of the production process. SPA provides a unique way of identifying processes in the entire supply chain with hot-spots thereby ensuring that appropriate intervention measures and effective policies can be prioritised and implemented to reduce carbon impacts. Emissions resulting from industries in the ROW indicate that biofuel energy policies should not be limited to the UK but rather a holistic approach should be adopted to account for emissions occurring beyond the boundaries of the UK. SPA for co-products was not undertaken since system expansion allocation was not used

As has been demonstrated for RME biodiesel, a systematic analysis of hybrid LCA and application of SPA should also be extended to second generation biofuel because the environmental impact of second-generation biofuel production can vary considerably depending on the conversion route as well as the feedstock and site-specific conditions ([50] and [51]). This is because, the benefits of second generation biofuels is being promoted (e.g. [1] and [52]), but the environmental profile is not fully understood.

Acknowledgement

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Supporting Information Available

Further tables (unit process exchanges, allocation and SPA results) and figures (biodiesel in the EU, biodiesel hybridised system boundary, results) and spreadsheet of input-output analysis are presented in the Supporting Information.

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Supporting Information (Environmental Science and Technology):

Identification of ‘Carbon Hot-Spots’ and Quantification of GHG Intensities in the Biodiesel Supply Chain using Hybrid LCA and Structural Path Analysis

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Figure S1: Timeline of Policies affecting Biofuel Production in Europe

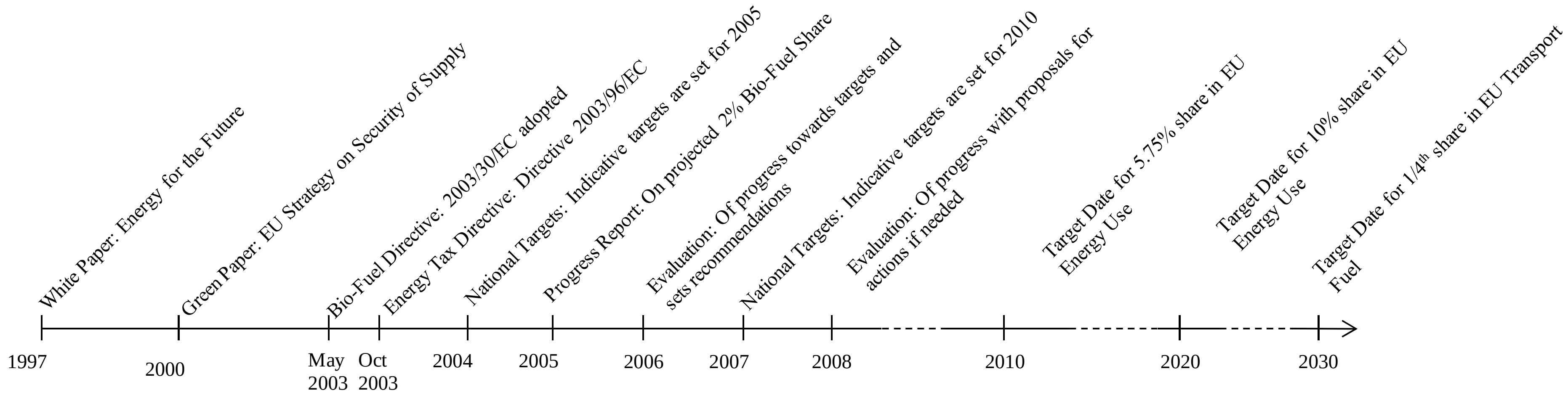

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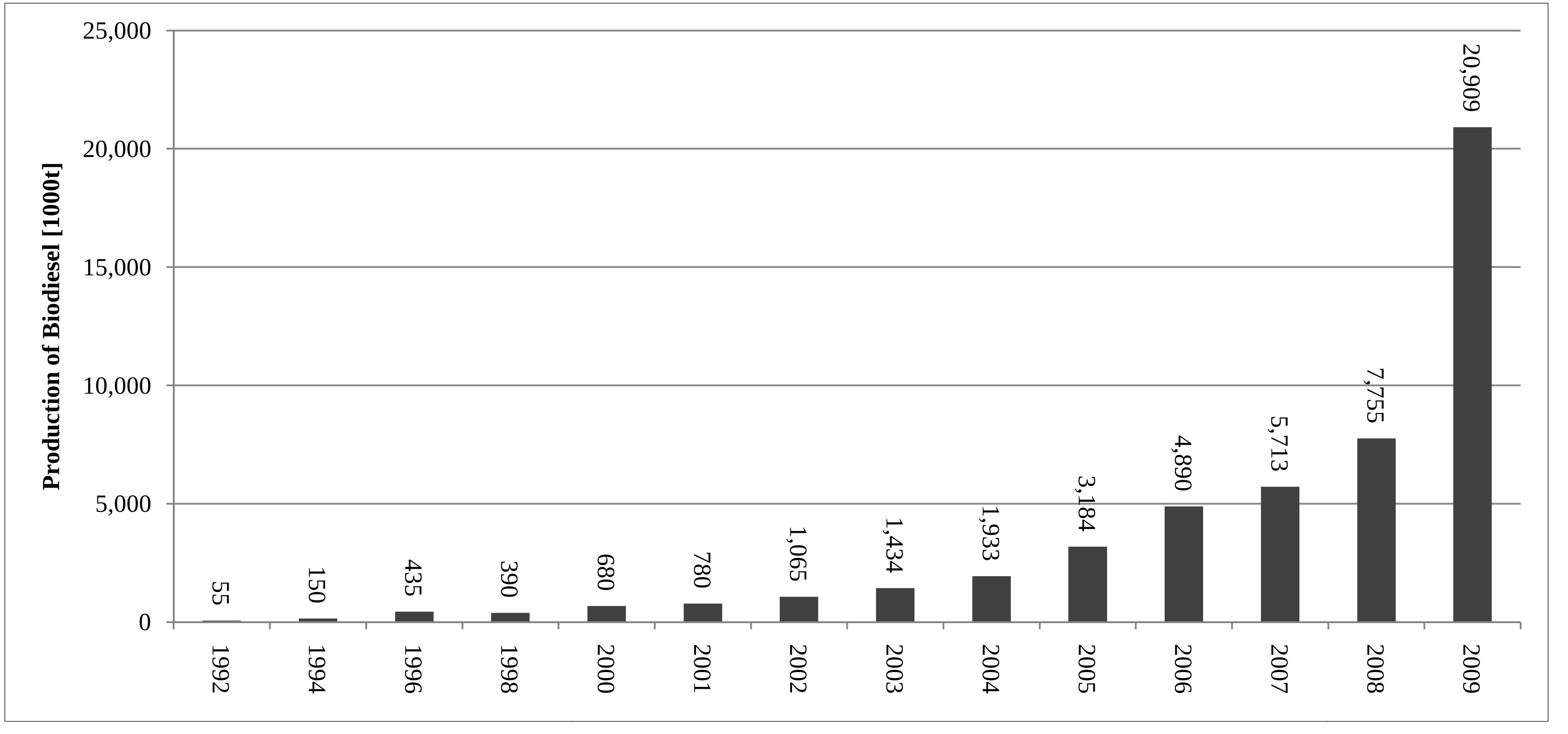
Figure S2: Trend in the growth of Biodiesel in Europe (Adapted from statistics of the European Biodiesel Board, 2009)



Reference: European Biodiesel Board, 2009. "Statistics: The EU Biodiesel Industry". Adapted from: http://www.ebb-eu.org/stats.php#

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Figure S3: Depiction of the Hybridised Systems Approach to GHG Assessment of Biodiesel Supply Chain used in this study

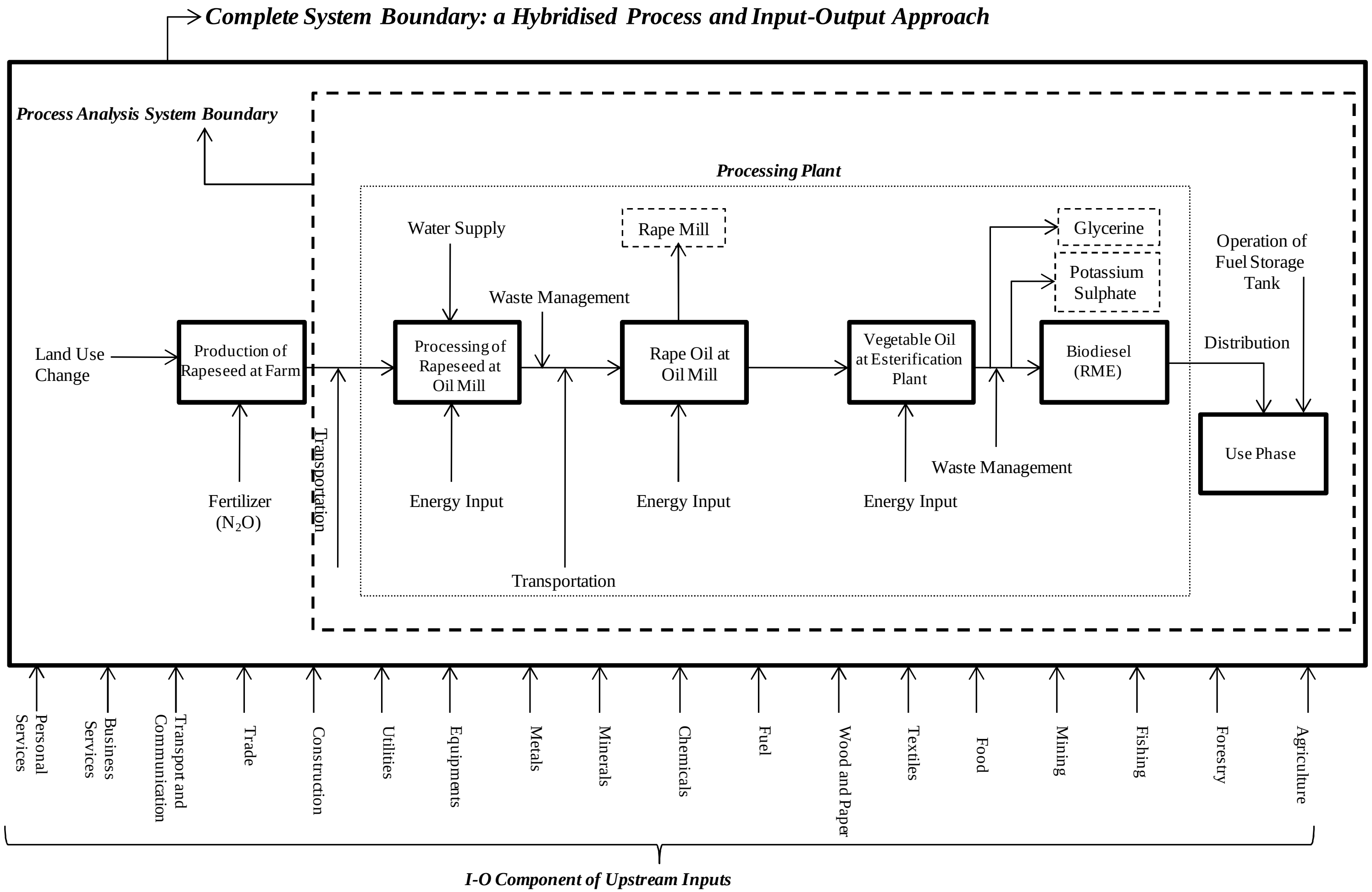

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Figure S4: Simplified representation of Structural Path Analysis of two products used in the process of producing biodiesel

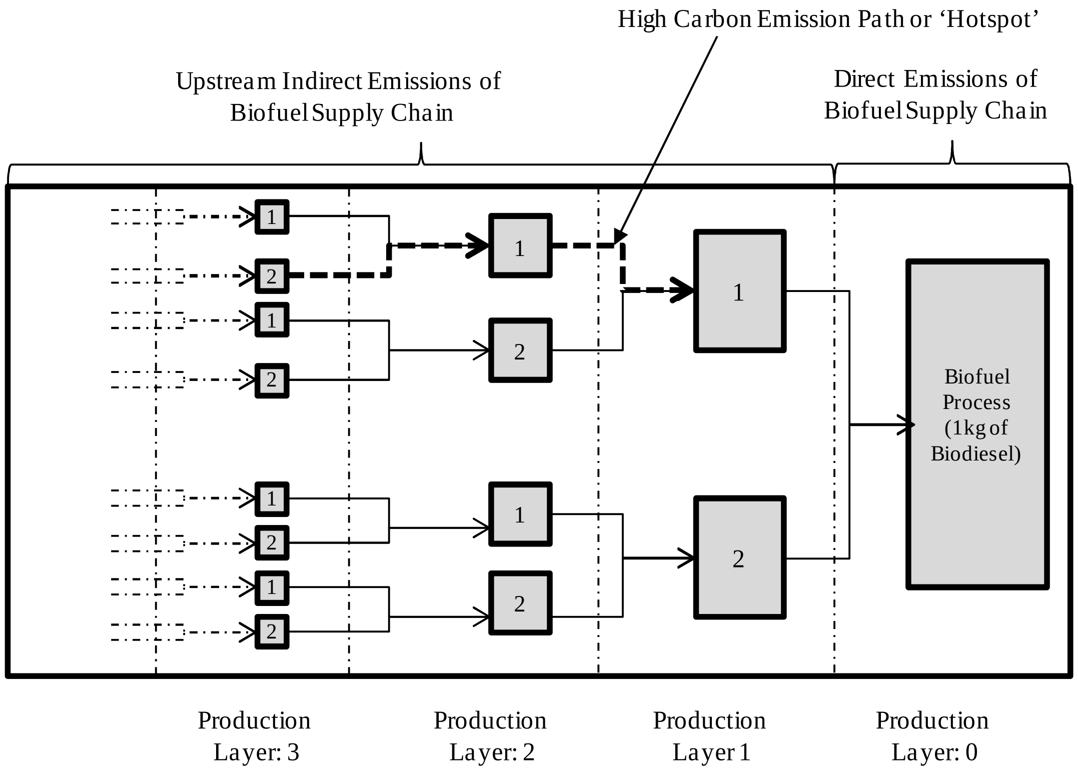

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Figure S5: Life Cycle Emissions of RME Production and Supply Chain normalized in energy units

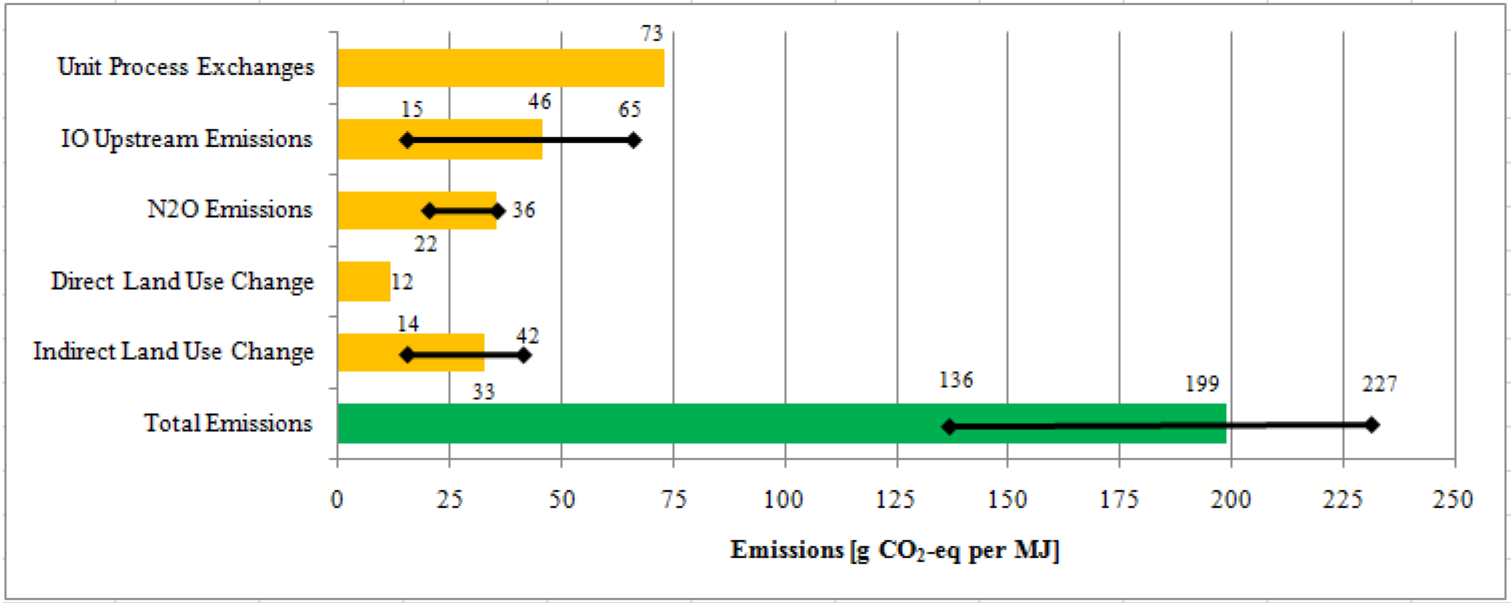

Figure S6: Depiction of build-up of environmental impact along higher upstream production layers in the biodiesel supply chain. The contributions of main sections of the economy are shown.

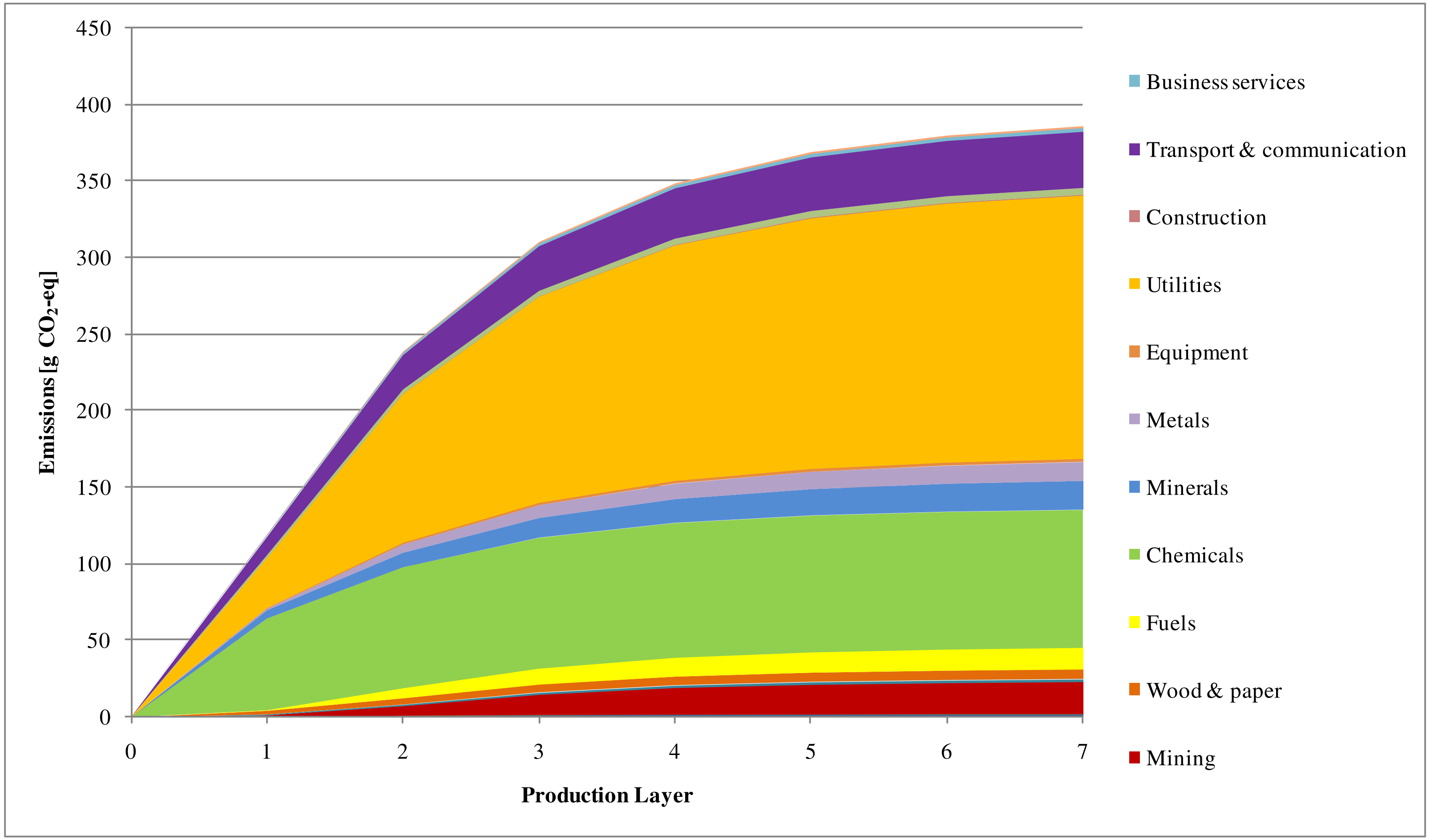

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Table S1: Description of Unit Process Exchanges for biodiesel supply chain

Unit Process Exchanges	***Description***
Biomass/Fuels: rape methyl ester production at esterification plant	The process includes the esterification process of oil to methyl ester, glycerine and potassium sulphate, intermediate storage of the oil and products, treatment of specific wastewater effluents. System boundary is at the esterification plant.
Electricity/Supply Mix: low voltage electricity generation at grid	Data set includes the transmission network infrastructure and emissions from transmission at low voltage
Oil/Heating System: light fuel oil burnt in 100kW non-modulating boiler	Processes include electricity use, waste and direct air emissions from combustion in the operation of operation of a light fuel oil boiler
Transportation Systems/Trains: transport, freight, rail	Inventory refers to the entire transport life cycle including production, maintenance and disposal, construction and maintenance and disposal of railway tracks.
Transportation System: transport, lorry >16t fleet average and transport, lorry 20-28t, fleet average	Inventory refers to the entire transport life cycle: operation of vehicle; production, maintenance and disposal of vehicles; construction and maintenance and disposal of road
Water supply/production: tap water, at user	Infrastructure and energy use for water treatment and transportation to the end user
Waste management/hazardous waste incineration: disposal, separator sludge, 90% water, to hazardous waste incineration	Waste-specific air and water emissions from incineration, auxiliary material consumption for flue gas cleaning. Short-term emissions to river water and long-term emissions to ground water from residual material landfill (from solidified fly ashes and scrubber sludge). Process energy demands for hazardous waste incineration
Waste management/sanitary landfill: disposal, municipal solid waste, 22.9% water, to sanitary landfill	Waste-specific short-term emissions to air via landfill gas incineration and landfill leachate. Burdens from treatment of short-term leachate (0-100a) in wastewater treatment plant
Waste management/wastewater treatment: class 2 wastewater treatment of rainwater from mineral oil storage and class 2 wastewater treatment of sewage	Infrastructure materials for municipal wastewater treatment plant transport, dismantling. Land use burdens

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Oil/Production: regional distribution, oil products	Infrastructure (materials and land use) for storage tanks and petrol stations. Bottom-Up estimation based on plant data. Life time is 80 years. Product storage volume of storage tanks is 10,000 m^3 with average storage time of 2 months

Table S2: Allocation Factor of RME Production Processes (Adapted from [24])

Production Stage	***Products***	***Allocation Factor (%)***
Oil Mill	Rape Oil	75.4
	Oil Mill	24.6
Esterification Plant	Rape Methyl Ester	86.9
	Glycerine	12.9
	Potassium Phosphate	0.2

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Table S3: Top-50 of the Ranked Structural Paths contributing to RME Biodiesel Upstream Supply Chain Emissions (UK = United Kingdom, ROW = Rest of World; see Supporting Spreadsheet S1 for numbering of sectors)

Rank	Supply Chain Path Description of RME Biodiesel	Path value [g CO_2-eq]	Path Order	Percentage in Total Impact
1	ROW-102 Other chemical products > Biofuel Process	20.70	1	5.35%
2	ROW-95 Organic basic chemicals > Biofuel Process	19.70	1	5.10%
3	UK-151 Electricity - coal > Biofuel Process	17.00	1	4.41%
4	ROW-161 Distribution and trade in electricity in electricity > ROW-102 Other chemical products > Biofuel Process	9.52	2	2.46%
5	ROW-161 Distribution and trade in electricity in electricity > ROW-95 Organic basic chemicals > Biofuel Process	9.09	2	2.35%
6	UK-152 Electricity - gas > Biofuel Process	6.59	1	1.71%
7	UK-182 Freight transport by road > Biofuel Process	5.56	1	1.44%
8	ROW-97 Plastics and synthetic rubber > Biofuel Process	4.65	1	1.20%
9	UK-186 Passenger air transport > Biofuel Process	3.46	1	0.90%
10	UK-116 Aluminium > Biofuel Process	3.39	1	0.88%
11	UK-97 Plastics and synthetic rubber > Biofuel Process	3.38	1	0.87%
12	UK-151 Electricity - coal > UK-161 Distribution and trade in electricity > Biofuel Process	3.28	2	0.85%
13	ROW-94 Inorganic basic chemicals > Biofuel Process	2.75	1	0.71%
14	ROW-160 Transmission of electricity > ROW-102 Other chemical products > Biofuel Process	2.33	2	0.60%
15	ROW-160 Transmission of electricity > ROW-95 Organic basic chemicals > Biofuel Process	2.23	2	0.58%
16	ROW-161 Distribution and trade in electricity > ROW-97 Plastics and synthetic rubber > Biofuel Process	2.15	2	0.56%
17	UK-94 Inorganic basic chemicals > Biofuel Process	1.84	1	0.48%

18	UK-170 Wholesale trade > Biofuel Process	1.80	1	0.47%
19	ROW-152 Electricity - gas > ROW-102 Other chemical products > Biofuel Process	1.73	2	0.45%
20	UK-152 Electricity - gas > UK-162 Gas distribution > Biofuel Process	1.72	1	0.44%
21	ROW-152 Electricity - gas > ROW-95 Organic basic chemicals > Biofuel Process	1.65	2	0.43%
22	ROW-151 Electricity - coal > ROW-102 Other chemical products > Biofuel Process	1.46	2	0.38%
23	ROW-151 Electricity - coal > ROW-95 Organic basic chemicals > Biofuel Process	1.39	2	0.36%
24	UK-80 Articles of paper > Biofuel Process	1.32	1	0.34%
25	ROW-105 Plastic plates, sheets > ROW-102 Other chemical products > Biofuel Process	1.25	2	0.32%
26	ROW-161 Distribution and trade in electricity > ROW-94 Inorganic basic chemicals > Biofuel Process	1.24	2	0.32%
27	UK-152 Electricity - gas > UK-161 Distribution and trade in electricity > Biofuel Process	1.24	2	0.32%
28	ROW-105 Plastic plates, sheets > ROW-95 Organic basic chemicals > Biofuel Process	1.20	2	0.31%
29	UK-182 Freight transport by road > UK-170 Wholesale trade > Biofuel Process	1.13	2	0.29%
30	ROW-95 Organic basic chemicals > UK-97 Plastics and synthetic rubber > Biofuel Process	1.10	2	0.28%
31	ROW-105 Plastic plates, sheets > Biofuel Process	1.04	1	0.27%
32	UK-105 Plastic plates, sheets > Biofuel Process	1.01	1	0.26%
33	ROW-100 Pharmaceuticals > ROW-102 Other chemical products > Biofuel Process	1.00	2	0.26%
34	ROW-100 Pharmaceuticals > ROW-95 Organic basic chemicals > Biofuel Process	0.96	2	0.25%
35	UK-95 Organic basic chemicals > Biofuel Process	0.90	1	0.23%
36	ROW-154 Electricity - nuclear > ROW-102 Other chemical products > Biofuel Process	0.90	2	0.23%
37	ROW-154 Electricity - nuclear > ROW-95 Organic basic chemicals > Biofuel Process	0.86	2	0.22%
38	UK-151 Electricity - coal > UK-161 Distribution and trade in electricity > UK-162 Gas distribution > Biofuel Process	0.84	3	0.22%
39	UK-151 Electricity - coal > UK-162 Gas distribution > Biofuel Process	0.81	2	0.21%
40	UK-157 Electricity by biomass > Biofuel Process	0.79	1	0.20%

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41	ROW-161 Distribution and trade in electricity > Biofuel Process	0.75	1	0.19%
42	ROW-85 Motor spirit (gasoline) > ROW-102 Other chemical products > Biofuel Process	0.71	2	0.18%
43	UK-180 Taxi operation > Biofuel Process	0.69	1	0.18%
44	UK-177 Inter-city coach sevice > Biofuel Process	0.68	1	0.18%
45	ROW-85 Motor spirit (gasoline) > ROW-95 Organic basic chemicals > Biofuel Process	0.68	2	0.18%
46	UK-151 Electricity - coal > UK-162 Gas distribution > Biofuel Process	0.67	1	0.17%
47	ROW-188 Supporting and auxiliary transport > ROW-102 Other chemical products > Biofuel Process	0.62	2	0.16%
48	ROW-113 Articles of concrete > ROW-102 Other chemical products > Biofuel Process	0.60	2	0.16%
49	UK-95 Organic basic chemicals. > UK-97 Plastics and synthetic rubber > Biofuel Process	0.59	1	0.15%
50	ROW-188 Supporting and auxiliary > ROW-95 Organic basic chemicals > Biofuel Process	0.59	2	0.15%