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

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16   **Abstract**

17   It is expected that biodiesel production in the EU will remain the dominant contributor as part

18   of a 10% minimum binding target for biofuel in transportation fuel by 2020 within the 20%

19   renewable energy target in the overall EU energy mix. Life cycle assessments (LCA) of

20   biodiesel to evaluate its environmental impacts have, however, remained questionable,

21   mainly because of the adoption of a traditional process analysis approach resulting in system

22   boundary truncation and because of issues regarding the impacts of land use change and N_2O

23   emissions from fertiliser application. In this study, a hybrid LCA methodology is used to

24   evaluate the life cycle CO_2 equivalent emissions of rape methyl ester (RME) biodiesel. The

25   methodology uses input-output analysis to estimate upstream indirect emissions in order to

26   complement traditional process LCA in a hybrid framework. It was estimated that traditional

27   LCA accounted for 2.7 kg CO_2-eq per kg of RME or 36.6% of total life cycle emissions of

28   the RME supply chin. Further to the inclusion of upstream indirect impacts in the LCA

29   system (which accounted for 23% of the total life cycle emissions), emissions due to direct

30   land use change (6%) and indirect land use change (16.5%) and N_2O emissions from fertiliser

31 applications (17.9%) were also calculated. Structural path analysis is used to decompose

32 upstream indirect emissions paths of the biodiesel supply chain in order to identify, quantify

33 and rank high carbon emissions paths or 'hot-spots' in the biodiesel supply chain. It was

34 shown, for instance, that inputs from the 'Other Chemical Products' sector (identified as

35 phosphoric acid, H_3PO_4) into the biodiesel production process represented the highest carbon

36 emission path (or hot-spot) with 5.35% of total upstream indirect emissions of the RME

37 biodiesel supply chain.

38

39 **1. Introduction**

40 There has been a growing interest in the use of biofuels as a sustainable replacement for fossil

41 fuels over recent years. This has led to many countries, including the UK and the wider EU

42 community, formulating policies that set out long-term strategies to promote biofuel

43 production and use driven mainly by policy goals such as: reducing greenhouse gas emissions

44 through the decarbonisation of transport fuels, diversifying fuel supply sources and

45 developing long-term replacements for fossil oil. The EU has a long-term vision for biofuels,

46 proposing that by 2030 and beyond, clean and CO_2-efficient biofuels would make up 25% of

47 the EU's transport fuel needs [1]. Refer to Figure S1 in the Supporting Information (SI) on

48 the web for an illustration of the transition plan of past EU policies affecting biofuels and the

49 timescale for future commitments.

50 Biodiesel is Europe's dominant renewable fuel [2] with rapeseed accounting for about 80%

51 of primary feedstock for biodiesel processing and about 75% share of total oilseed production

52 of EU-27 in 2009-10 [3]. Production of biodiesel on an industrial scale began in 1992 about

53 five years before the EU's Energy White Paper 'Energy for the Future', driven mainly by

54 positive signals in terms of support from member states and the EU Commission. Figure S2

55 under SI on the web shows the trend in the growth of biodiesel in the EU since 1992.

56 Despite the potential growth and benefits of biofuels, many research findings have raised

57 arguments against them in a global context. The Food and Agricultural Organisation [4]

58 acknowledges that, although biofuels under certain conditions help reduce greenhouse gas

59 emissions (GHG), the global effects of an expansion of biofuel production will depend

60 crucially on where and how feedstocks are produced. It is therefore anticipated that the more

61 sustainable second-generation biofuels produced from non-food crops and residues can

62 provide the best opportunity for the commercial viability and development of the sector from

63 2014 onwards [5]. Lifecycle assessment (LCA) of second-generation bioethanol produced

64 from surplus forest-bioenergy resources in Norway for example was estimated to potentially

65 save 6-8% of Norway's global warming GHG emissions associated with road transportation

66 [6]. The accelerating use of biomass including, cereals such as wheat, maize, sugar and

67 oilseed for biofuel and power generation has come about because of positive government

68 directives and political decisions [7]. However, the exact impact on the resource base and the

69 environment due to the demand for biofuels is unknown.

70 Many authors have therefore undertaken studies to evaluate the environmental impacts of

71 biofuel production [8-10]. These studies have mainly used traditional LCA methods based on

72 ISO 14040 and mostly involved comparative studies with traditional fossil fuel production

73 [11]. Traditional LCA of biofuel production involves setting a system boundary for the

74 biofuel supply chain and using process analysis data to estimate the carbon impacts of

75 selected supply chains within the system boundary.

76 It is, however, well recognised that because of difficulties in collecting process-specific data

77 in LCA and the infinite number of possible supply chain paths, the use of hybrid LCA

78 provides a more comprehensive framework for the evaluation of environmental impacts of

79 upstream production [12-15]. A hybrid LCA combines the specificity of process analysis

80 with the extended system boundary of input-output (IO) analysis. Hybrid LCA has had many

81 applications. Lenzen and Wachsmann [16] and Crawford [17] demonstrated the use of a

82 hybrid LCA technique in the assessment of the energy content of wind turbines in order to

83 achieve system completeness. A limited number of studies using hybrid LCA have been

84 undertaken on biofuels. Bright et al. [18] undertook an environmental assessment of wood-

85 based biofuel to estimate the cumulative global warming mitigation under different scenarios

86 in Norway. The hybrid LCA in this study consisted of a two-region (Norway and the

87 European Union) IO model and process analysis inventory for the biofuel options.

88 In this paper the life cycle GHG emissions of a typical biodiesel supply chain are calculated

89 using hybrid LCA, incorporating process-specific data of rape methyl ester (RME)

90 production and inputs from higher upstream processes such as chemical inputs, mining,

91 transportation, banking, equipment, etc, based on input-output analysis. Direct and indirect

92 emissions in the biofuel supply chain are determined, including direct and indirect land use

93 change and N_2O emissions from fertiliser application. Furthermore, structural path analysis (a

94 decomposition technique used in economic and ecological systems analysis) is applied to

95 identify, quantify and rank high carbon emission paths – or 'carbon hot-spots' – in the supply

96   chain. Some studies have demonstrated the use of SPA as a decomposition technique of

97   environmental impacts in a LCA context [19-20]. To the best of the authors' knowledge, this

98   is the first time that SPA has been used for an analysis of the biodiesel supply chain. This

99   detailed analysis is aimed at helping to tailor and prioritise mitigation efforts through the use

100   of biofuels.

101

102

103   **2. Material and Methods**

104   'Integrated hybrid LCA' as defined by Suh and Huppes [21] is applied in this study. This form

105   of hybrid LCA combines a process matrix and an IO matrix in a consistent mathematical

106   framework [22]. Whereas the process component systematically computes physical inputs

107   and outputs of each production step within the system boundary, the input-output component

108   completes the analysis by enumerating upstream indirect inputs from outside the process

109   system boundary.

110   For an integrated hybrid assessment of biofuel supply chains, the process matrix is linked to

111   the input-output matrix using the operational expenditure of biofuel production to account for

112   upstream inputs. As shown by Suh and Huppes [21], the general relationship for the

113   integrated hybrid model is given in matrix notation by:

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

115   Where:

116    $\widetilde{P}_{hybrid}$ = Total (direct and indirect) environmental impact (e.g. CO_2-eq emissions) associated

117   with one unit of final demand y for the product (here biodiesel).

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

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

120   I = identity matrix (dimension: $m \times m$)

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

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

123   E_p = process inventory environmental extension matrix. CO_2-eq emissions are

124   diagonalised (dimension: $m \times s$)

125   E_{i-o} = IO environmental extension matrix. CO_2-eq emissions are diagonalised

126   (dimension: $m \times s$)

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

128   except y

129   Matrix A_p describes the product inputs into processes as captured in the unit process

130   exchanges (or process analysis inventory from ecoinvent in this case) and described in Table

131   S1. These processes, together with the sectoral inputs from IO sectors, are used to draw up

132   the biodiesel supply chain map as depicted in Figure S3.

133   Matrix U, which is assigned a negative sign, represents the higher upstream inputs from the

134   IO system to the process system. Matrix D, also assigned a negative sign, represents the

135   (downstream) use of goods / process inputs from the process to the background economy (IO

136   system). As explained by Suh and Huppes [21], the downstream cut-off matrix represents the

137   link from the process-based (foreground) system to the IO-based (background) system. It can

138   be argued that the downstream cut-off flows in D are often small compared to the – normally

139   much larger – background economy (cf.[23]). The aim of the present paper is to quantify the

140   total emissions of biodiesel production in the status-quo economy of the UK in 2004 when

141   the market share of biodiesel as a percentage of road transport fuel was at a modest 0.09% in

142   2004 [24]. For the sake of simplification we therefore neglect interactions with the

143   background economy and set values in D are set to zero. We acknowledge, however, that a

144   more general use of biodiesel in the economy would ideally be evaluated by including

145   industries' expenditures on biodiesel in D (e.g. by assuming different market penetrations of

146   biodiesel in a number of scenarios).

147   The final demand $\tilde{y}$ for biodiesel also represents the functional unit of the LCA system, set to

148   1kg of RME biodiesel in this study.

149   In order to achieve a complete LCA system for the biodiesel supply chain, upstream cut-offs

150   from the process-based LCA system were estimated using input-output analysis. For

151   example, to estimate the contributions of an upstream service (for example: administration)

152   for a given process inventory (for example: electricity) already captured in the process

153   matrix, $\mathbf{A_p}$ the following steps were taken. The unit cost of the process under consideration

154   (example: electricity) was obtained [£/kWh]. This was multiplied by the input (in physical

155   terms) of electricity [kWh] obtained from the process matrix. The results, k (that is:

156   [£/kWh]* [kWh]) represents the amount of electricity (in £) needed to produce 1kg of final

157   demand of biodiesel. This amount is then used as a scalar multiplier to the column a_{ij} of the

158   IO technology matrix, A_{i-o} where j corresponds to the Electricity industry. To avoid double

159   counting, all inputs already captured in the process matrix are discounted from the resulting

160   column vector ka_{ij}. The corrected values $ka_{ij}*$ become elements of the upstream input

161   matrix U. The administrative expenditure linking the process LCA electricity to the IO table

162   corresponds to ka_{ij} where i corresponds to Administration as a product and j Electricity as

163   an industry. Refer to Spreadsheet S1.

164   Uncertainty in upstream emissions was estimated by including the maximum/minimum IO

165   upstream cut-offs into the LCA system. To account for the maximum IO upstream cut-offs,

166   all potential sectoral products that are indirect input requirements into biodiesel production

167   are included. Similarly, to account for minimum IO upstream cut-offs, only sectoral products

168   that are highly probable indirect input requirements into biodiesel production supply chain

169   are included. Refer to the supplementary Spreadsheet S1 for inputs into the upstream supply

170   chain for the maximum and minimum case scenarios. Besides its mathematical consistency,

171   integrated hybrid LCA provides a comprehensive framework because all inputs associated

172   with the biodiesel supply chain can be expressed by the combination of process and IO

173   matrices.

174

175   **2.1 Structural Path Analysis**

176   Taylor's series expansion is applied only to the IO part of Equation (1) because the inputs of

177   the unit process obtained from ecoinvent are clearly known:

178 $$P'_{IO} = [E] \times (I - A)^{-1}y = \mathbf{EIy} + \mathbf{EA^1y} + \mathbf{EA^2y} + \mathbf{EA^3y} + \cdots + \mathbf{EA^n y} \qquad \text{(Equation 2)}$$

179 EIy', represents the direct GHG emissions emitted (at production level 0) for a given demand

180 and $EA^n y$ the indirect GHG emissions emitted for a given final demand at the nth production

181 level. The Taylor series expansion of the Leontief inverse matrix can be further decomposed

182 by unravelling the A matrix (or the IO technology coefficient matrix with elements, a_{ij}) into

183 a series of structural paths at the nth order to systematically identify important supply chains

184 [25]. Summing up across all products i and industries, j, the total environmental impact of a

185 final demand bundle y_i can be decomposed into: (Equation 3)

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

186

187 where e_j is the emission intensity of industry j and elements a_{nm} represent transaction

188 coefficients between sector n and m. Each multiplied term represents the contribution of an

189 individual supply chain path. In the case of biodiesel in this study, the emission 'carbon hot

190 spots' in the supply chain were to be identified and therefore the combined upstream inputs

191 from matrix U as the demand bundle y_i were used: (Equation 4)

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

192

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

194   The decomposition of the series expansion can be represented as a tree diagram (Refer to

195   Figure S4) whereby each tier in the tree represents a different production layer and each node

196   gives the contribution to total environmental impacts from the demand, y [26].

197   Production layer refers to the stage of supply to the main product. Production layer 0

198   therefore refers to the biodiesel production process. Production layer 1 is the first stage of the

199   upstream supply chain and production layer 2, is the supplier to the first upstream supplier of

200   the biodiesel process. In a SPA disaggregation of a product system, each node represents a

201   contribution to the total environmental impacts from the demand, y.

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

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

204   n = Number of sectors in the economy

205   l = Production level

206   The importance of supply chain decomposition in disentangling upstream emission paths in a

207   product system is evident in the fact that upstream environmental impacts are often greater

208   than direct environmental impacts in a supply chain. In a carbon footprint case study of

209   economic sectors in Australia and the US, Huang et al. [27] for example, showed that direct

210   emissions of the majority of sectors are below 20% of the total carbon footprint, and can be

211   as low as 1%. To maximise the potential for biodiesel to achieve real CO_2 emissions

212   reductions compared to fossil fuels, high emissions intensity paths or 'hot-spots' in the supply

213   chain must be identified and possible lower emission alternative processes for the production

214   of biofuels must be found.

215

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.

232

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].

236   Wiedmann et al. [29] describe the construction of a multi-regional input-output (MRIO)

237   model using UK national IO tables and rest-of-world (ROW) tables from the Global Trade

238   Analysis Project (GTAP). A technology coefficients matrix was derived for both the UK

239   domestic and UK imports use table. For the purpose of the present study, the ROW economy

240   is represented as one symmetric table (technical details of this 2-region model have been

241   described in [30].

242   Columns of UK and ROW industry input requirements are augmented with data for

243   greenhouse gas emissions to derive sectoral emissions intensities (kg CO_2-eq/£) for the

244   environmental matrix, E_{i-o}. A supply chain map illustrating the comprehensive system

245   boundary framework of the biodiesel supply chain adopted in this study is available in Figure

246   S3 under SI on the web.

247

248   **2.4 Allocation Factors**

249   The production of RME results in multiple product outputs. For example, the processing of

250   oil mill into rape oil also results in the production of rape mill as a by-product. The

251   esterification of vegetable oil into RME also produces glycerine and potassium sulphate. In

252   order to deal with multiple product outputs, LCA studies apply the method of either

253   allocation or system expansion. In the first case, inventory data are allocated to the main

254   product, by-products and waste, respectively, in order to assign material inputs and

255   environmental impact. In system expansion, the boundary is extended to account for the input

256   and output flows of all products. In this study, we use the first option, allocation, as we are

257   specifically interested in the provision of biodiesel.

258   Allocation factors can be based either on mass flow, energy value or economic revenue of co-

259   products. Economic allocation has been established as a recognised way of systematically

260   executing allocation in LCA [31-33]. The International Standards Organisation [34] also

261   gives this allocation option in Step 3 of its allocation procedure. Hence, in this study, the

262   economic revenue allocation as adopted in ecoinvent [28] was used. To reduce the

263   uncertainty related to economic allocation because of potential fluctuations in the economic

264   values of product and co-products, the environmental burdens are allocated according to the

265   revenue of all process products, based usually on the average prices for three consecutive

266   years. (Refer to Table S2 for allocation details). Allocation factors for other methods related

267   to the production of RME are also presented in ecoinvent [28].

268

3. Results and Discussions

3.1 Hybrid Life Cycle CO_2-eq of Biodiesel Production

271   The total emissions of all unit process exchanges representing the process analysis data of the

272   biodiesel production process is 2.7 kg CO_2-eq or 36.6% of the total life cycle emissions. IO

273   upstream indirect emissions (for the base case impact scenario) account for 1.7 kg CO_2-eq of

274   the total life cycle emissions. Upstream emissions include embodied emissions such as those

275   associated with utilities, equipments, chemicals, mining, construction of buildings,

276   maintenance, services such as banking and finance, insurance, research and development,

277   advertising, etc., and accounted for approximately 23% of total emissions. A further

278   breakdown of these emissions is provided in Section 3.3 (Structural Path Analysis of

279   Biodiesel Supply Chain Emissions). Refer also to Table S3 and Figure S6 on the web.

280   It was also estimated (from the process analysis inventory in ecoinvent) that the esterification

281   of vegetable oil to RME process accounted for 35.5% of the total emissions or 97% of

282   emissions due to the unit process exchanges in the process inventory. The other unit process

283   exchanges: road and train transport, electricity supply, regional distribution of oil, waste

284   management and water treatment from the process analysis inventory in ecoinvent

285   collectively accounted for around 1.1% of the total emissions associated with the RME

286   biodiesel production process.

287

288   **3.2 Other Impacts**

289   It has generally been argued that greenhouse gas releases from land use change and nitrous

290   oxide (N_2O) emissions from the use of fertilisers can potentially be significant enough to

291   change the environmental profile of biodiesel [35-36].

292   With N_2O having a global warming potential 298 times that of CO_2 when considered over a

293   100-year period [37], the use of nitrogen fertilisers has the potential to significantly affect the

294   GHG emissions balance of biodiesel. N_2O is emitted both directly from soils due to the use of

295   nitrogen-based fertilisers and microbial transformations of organic nitrogen (N), and also

296   indirectly with nitrogen losses through volatilization, leaching and runoff of N-compounds

297   that are converted into N_2O off site.

298   Also, the European Commission Joint Research Centre [38], Searchinger et al. [39] and

299   Fargione et al. [40] have all stated that indirect land-use change could potentially release

300   enough greenhouse gases to negate the savings from conventional biofuels. Land use can be

301   defined as the type of activity being carried out on a unit of land and the change in land use

302   can be either direct or indirect. Direct land-use change occurs when feedstock being

303   cultivated for biofuels production (e.g. rapeseed for biodiesel) displaces a prior land use (e.g.

304   forest), thereby generating possible changes in the carbon stock of that land. Indirect land-use

305   change on the other hand occurs when pressure on agriculture due to the displacement of

306   previous activity or use of the biomass induces land-use changes on other land [11].

307

308   **3.2.1 Estimation of N_2O Emissions from Fertiliser Application**

309   The Intergovernmental Panel on Climate Change, IPCC [41] estimate that the direct emission

310   factor associated with N_2O emissions is likely to be 1% of the N applied to the soil. Jungbluth

311   et al. [28] also estimated a direct emissions factor of 1.25% of the N-input and an indirect

312   emission factor of 2.5% from the nitrogen that is leached as nitrate. Crutzen et al. [42]

313   however state that global analysis of N_2O emissions had been previously underestimated and

314   shows that N_2O emission factor (direct and indirect) in agro-biofuel production is 3–5% of N

315   applied (that is 0.03-0.05 kg N_2O-N (kg N)$^{-1}$. The maximum value of 5% in Crutzen et al.

316   [42] is used to assess the highest impact case scenario in this study. The minimum value of

317   3% Crutzen et al. [42] is applied to determine the uncertainty range between the maximum

318   and the minimum impact scenario. Refer to Smeets et al. [43] for additional information on

319   the contribution of N_2O to the greenhouse gas balance of first-generation biofuels. The

320   fertilizer input rate assumed in this study (137.4 kg-N ha^{-1} yr^{-1}) was used in the JEC - Joint

321   Research Centre-EUCAR-CONCAWE collaboration on biofuel programme [44].   In the

322   cultivation of 1kg of rape, the following conditions were assumed:

323   i.   Land Use: Transformation from non-irrigated arable land accounted for 71% of land

324   use at 2.08 m^2 per kg of rape while transformation from pasture and meadow land

325   accounted for 29% of land use at 0.85m^2 per kg of rape [45]

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

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

328   of RME biodiesel

329

330   The relationship used to determine the life cycle CO_2-eq emissions associated with the use of

331   nitrogen fertilizers can be expressed as: Equation 6

$$\text{Life Cycle Emissions due to N}_2\text{O Use} = e_f y f_i r_s k_n (GWP)_{N_2O} \sum_{i=1}^{j} l_i p_i$$

332   Where:

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

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

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

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

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

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

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

340    $p =$ ratio of the area of particular land type occupied by bioenergy crop to the total area of

341   different land used (in cases where only one land type is used, p will be 1)

342

343 Using a GWP of 298 for N_2O and effective land size of 1.723 m^2 or $(1.723 \times 10^{-4}$ ha) per

344 kg of rape seed, it is estimated from Equation 6 that the N_2O emissions contributed between a

345 high impact scenario of 1.32 kg CO_2-eq per kg of RME and a low impact scenario of 0.80 kg

346 CO_2-eq per kg of RME biodiesel.

347

348 **3.2.2 Direct and Indirect Land Use Change**

349 The Intergovernmental Panel on Climate Change [47] reports that due to direct land use

350 change, changes in carbon stock per hectare of bioenergy crop cultivation occurs in the

351 following carbon pools for cropland: biomass (above ground biomass and below ground

352 biomass); dead organic matter (dead wood and litter) and soils (soil organic matter). The total

353 change in carbon stock is calculated using Equation 7 below [10].

354 Equation 7:

355 $\Delta \text{ Carbon Stock Soil } \left[\frac{tC}{ha}\right] = \sum_i \text{Carbon Stock Change Factor } \left[\frac{tC}{ha.yr}\right] \times T_i[yr]$

356 Where

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

358 Based on data on carbon stock change factors for the carbon pools of cropland from the IPCC

359 Guidelines for National Greenhouse Gas Inventories – Agriculture Forestry and other Land

360 Use [47], direct land use change was estimated to be 0.44 kg CO_2-eq per kg of RME.

361 Indirect land use change (iLUC) is calculated using the theoretical global average indirect

362 land use change factor [48]. In this study it was assumed that the cultivation of rape seed

363   occurred on 71% arable and 29% pasture and meadow land. A 'maximum risk' or 'maximum

364   iLUC order of magnitude' representing a 75% share of non-zero risk biofuel is assigned an

365   iLUC factor of 15 t CO_2-eq/ha/yr while a 'low risk' or 'low iLUC order of magnitude'

366   representing 25% of all non-zero risk biofuels are subject to theoretical full iLUC factor of 5 t

367   CO_2-eq/ha/yr [48]. The term risk refers to the level of impact due to the conversion of food

368   crop land into bioenergy crop land. A low risk biofuel is therefore assumed to be produced

369   from feedstock cultivated on set-aside or abandoned land. By weighting these iLUC factors

370   according to the ratio of land type and land sizes assumed in the cultivation of rape seed in

371   this study, the iLUC factor used for to the production of 1 kg of RME biodiesel and its co-

372   products is estimated to be 11.8 t CO_2-eq/ha/yr or 1.86 kg CO_2-eq. Taking into account the

373   allocation factors in Table 2, iLUC is calculated to be 1.22 kg CO_2-eq per kg of RME.

374   Uncertainty in the impact of land-use change refers to the variability of indirect land-use

375   change factors due to the type of land used in the cultivation of feedstock. Based on the

376   assumptions for maximum/minimum iLUC risk referred to above, the uncertainty range for

377   iLUC for producing 1kg of RME biodiesel is estimated to be between of 0.52 to 1.55 kg CO_2-

378   eq.

379   The emissions associated with all stages of the RME biodiesel production are shown in

380   Figure 1. The total life cycle CO_2-eq emissions for 1kg of RME biodiesel were calculated to

381   be 7.38 kg CO_2-eq or 199 g CO_2-eq/MJ. By accounting for uncertainty in the assessment, it

382   was estimated that the results are in the range 5.03 to 8.44 kg CO_2-eq per kg of RME

383   biodiesel. Refer to Figure S5 for normalized results in energy units.

384

385   **Figure 1:** Life Cycle Emissions of RME Production and Supply Chain
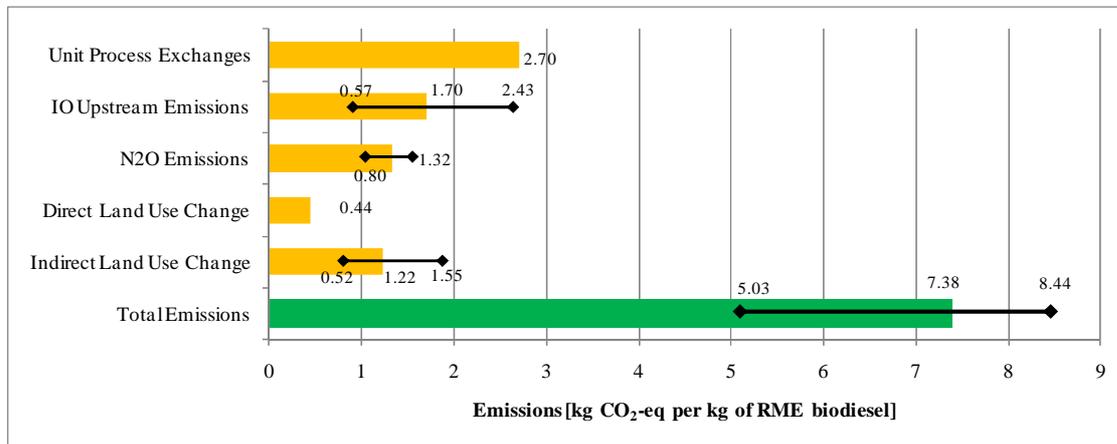
386

387   **3.3 Structural Path Analysis and Hotspots of Biodiesel Supply Chain Emissions.**

388   In Figure S6, the cumulative impacts of sectoral emissions from the higher upstream supply

389   chain paths of biofuel production are presented giving an indication of the relative

390   contribution of each IO sector. These higher upstream supply chain paths represent the IO

391   component of upstream inputs. Seven production layers of RME biodiesel were analysed. It

392   was estimated that the 'Utilities Sector' was the highest sectoral emitter accounting for 172 g

393   CO_2-eq or 44.5% of total upstream emissions. This was followed by the 'Chemical Sector'

394   emitting 90 g CO_2-eq or 23.3% of total upstream emissions. As can be seen from Figure S6,

395   the next four sectoral emissions were 'Transportation and Communication' (37 g CO_2-eq or

396   9.6%), 'Mining' (21 g CO_2-eq or 5.4%), 'Minerals' (19 g CO_2-eq or 4.8%) and 'Fuels'

397   (14 g CO_2-eq or 3.7%).

398   Structural path analysis (SPA) is used to show the inter-connections of various products and

399   industries within the biodiesel supply chain and identify, rank and estimate the CO_2-eq of the

400   high emissions intensity paths or 'carbon hot-spots'. 150 of the most important paths of the

19

401 biodiesel supply chain were extracted in the SPA. The cut-off threshold for individual path

402 contributions was set at 0.05% of total impacts in the analysis of the supply chain paths.

403 Detailed results for each of the top 50 paths are shown in Table S3 under SI on the web.

404 It was found that CO_2-eq emissions impacts on the biodiesel supply chain originate across the

405 entire economy but of the top 150 paths, the majority originate from the sectors 'Other

406 Chemical Products', 'Organic Basic Chemicals', 'Electricity-Coal', 'Distribution and Trade',

407 'Electricity-Gas' and 'Freight transport by Road'.

408 The "hottest spot" or the highest carbon intensity path of the biodiesel upstream supply chain

409 was identified as a path order 1: Rest of World (ROW) Sector (102) 'Other Chemical

410 Products' > Biofuel Process with an estimated 20.7 g CO_2-eq or 5.35% of the total emissions.

411 This path describes the emissions chain: 'Other Chemical Products' used as an input in the

412 biodiesel production process.

413

414 **4. Discussion**

415 The life cycle assessment of the RME biodiesel supply chain estimated the total life cycle

416 emissions of biodiesel production to be 7.38 kg CO_2-eq per kg with an uncertainty range of

417 5.03 to 8.44 kg CO_2-eq per kg. The uncertainty was a result of variability in indirect land use

418 change, N_2O emissions and IO higher upstream emissions. IO higher upstream emissions

419 accounted for approximately 23% of total CO_2-eq emissions. The use of a hybrid method

420 ensured the integration of process and IO analysis such that higher upstream inputs into

421 sectors such as utilities, transportation, chemicals, mining, services, etc which are normally

422 excluded from traditional life cycle assessments, are taken into account. In contrast, Halleux

423 et al. [8], Hoefnagels et al. [10] and Kim et al. [49] all undertook life cycle assessments of

424 biofuels using traditional LCA but did not account for upstream emissions outside the process

425 system boundary resulting in the truncation of the product system. Given that some past

426 assessments of biofuels have also neglected the impacts of land use change and N_2O

427 emissions, assuming that the impacts of land use change, N_2O emissions and IO upstream

428 emissions cut-offs were truncated from the biofuel product system, as has previously been the

429 case, the total emissions would have been 2.7 kg CO_2-eq per kg. This would have resulted in

430 a 63% underestimation of the total life cycle emissions. Therefore, IO upstream emissions

431 cut-off, N_2O emissions and land-use change represent significant impacts which are

432 determinants that can change the environmental profile of the biodiesel supply chain. It was

433 observed that the lack of process-specific data increases uncertainties in life cycle

434 assessments. The uncertainty estimates for this study were based on data variability in

435 indirect land use change, N_2O emissions from fertiliser application and aggregation of IO

436 data, resulting in the estimation of minimum and maximum carbon impacts of the RME

437 biodiesel supply chain. The estimation of emissions was based on economic allocation

438 between the RME biodiesel supply chain and co-products. This has been recognised as one

439 way of systematically executing allocation in LCA [31-33].

440 Structural path analysis (SPA) is useful in describing and characterizing carbon hotspots in

441 the supply chain. Specific processes in the RME biodiesel supply chain can be matched to the

442 structural paths in order to identify the hotspots in the supply chain. For example, the first

443 ranked structural path: Other chemical products > Biofuel Process can be identified as the

444 inputs of industrial grade phosphoric acid, H_3PO_4 (85% in water) into the biodiesel

445 production process. Likewise, the second ranked structural path: Organic basic chemicals >

446 Biofuel Process describes the inputs of methanol into the biodiesel production process. As

447 can be seen from the ranked structural paths available in the Table S3, all the paths end as a

448 direct input into the RME diesel production process. The demanding sector is therefore

449 responsible for the emissions caused, but the emissions might occur upstream of that sector.

450 For example, in the 12th ranked structural path: UK- Electricity - Coal> UK- Distribution and

451 Trade in Electricity > Biofuel Process; the biodiesel production process is responsible for

452 emitting 3.28 g CO_2-eq per kg of RME biodiesel although it occurs upstream of the

453 production process. SPA provides a unique way of identifying processes in the entire supply

454 chain with hot-spots thereby ensuring that appropriate intervention measures and effective

455 policies can be prioritised and implemented to reduce carbon impacts. Emissions resulting

456 from industries in the ROW indicate that biofuel energy policies should not be limited to the

457 UK but rather a holistic approach should be adopted to account for emissions occurring

458 beyond the boundaries of the UK. SPA for co-products was not undertaken since system

459 expansion allocation was not used

460 As has been demonstrated for RME biodiesel, a systematic analysis of hybrid LCA and

461 application of SPA should also be extended to second generation biofuel because the

462 environmental impact of second-generation biofuel production can vary considerably

463 depending on the conversion route as well as the feedstock and site-specific conditions ([50]

464 and [51]). This is because, the benefits of second generation biofuels is being promoted (e.g.

465 [1] and [52]), but the environmental profile is not fully understood.

466

Supporting Information Available

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

478

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612

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

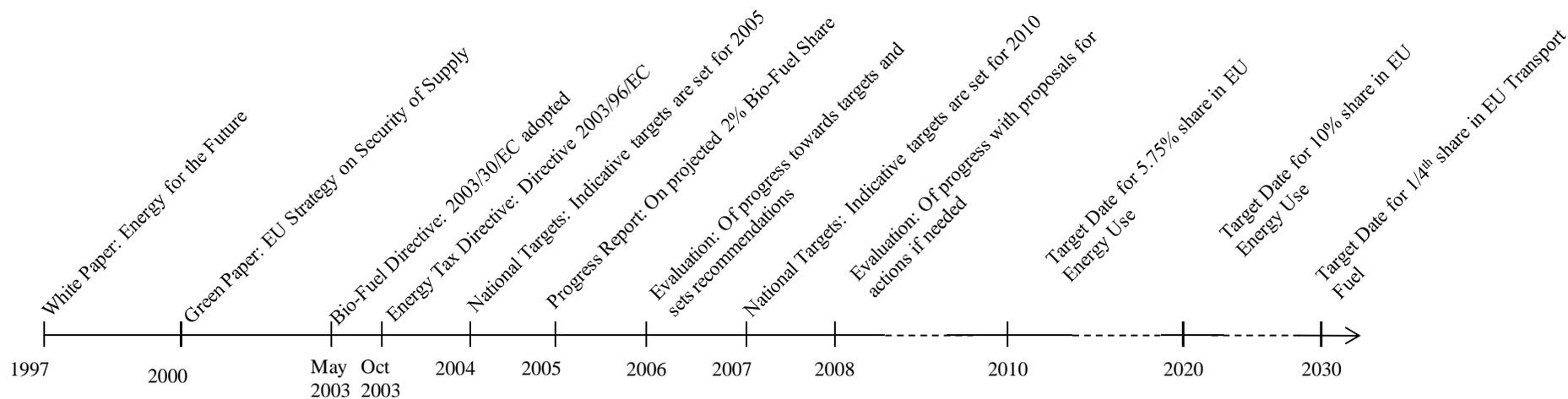**First Author** and Corresponding Author
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Figure S1: Timeline of Policies affecting Biofuel Production in Europe

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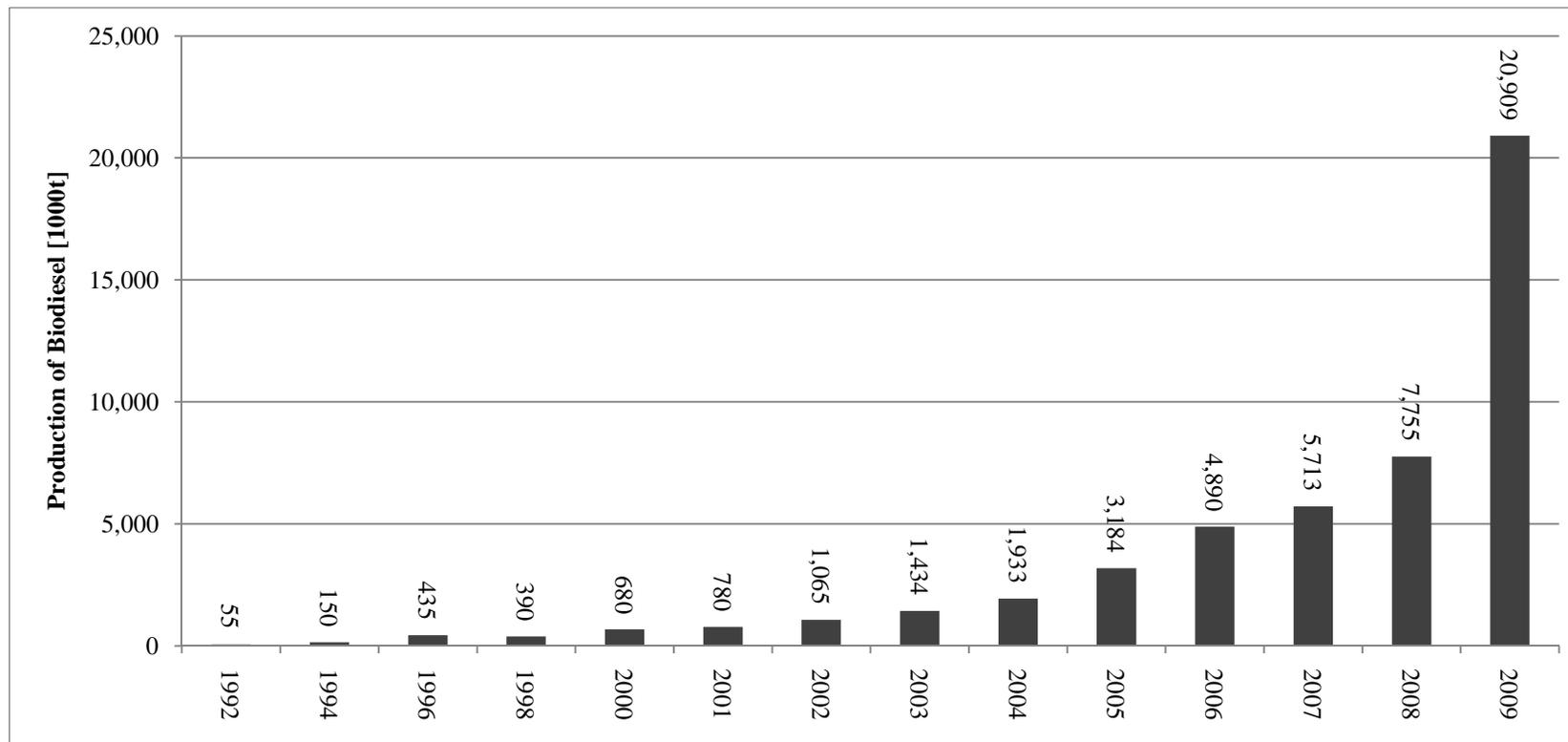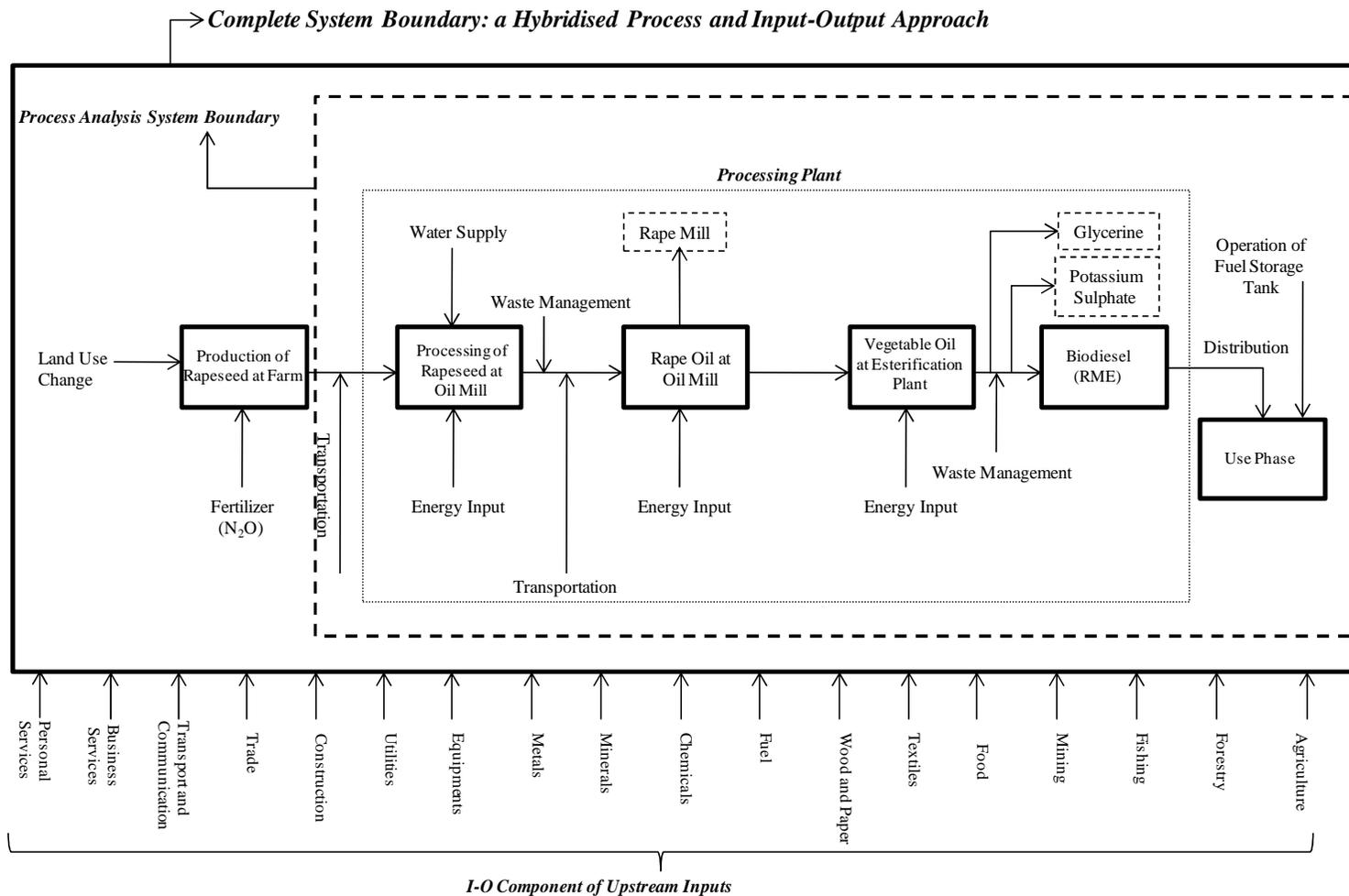

**Reference:** European Biodiesel Board, 2009. "Statistics: The EU Biodiesel Industry". Adapted from: http://www.ebb-eu.org/stats.php#

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

Figure S4: Simplified representation of Structural Path Analysis of two products used in the process of producing biodiesel
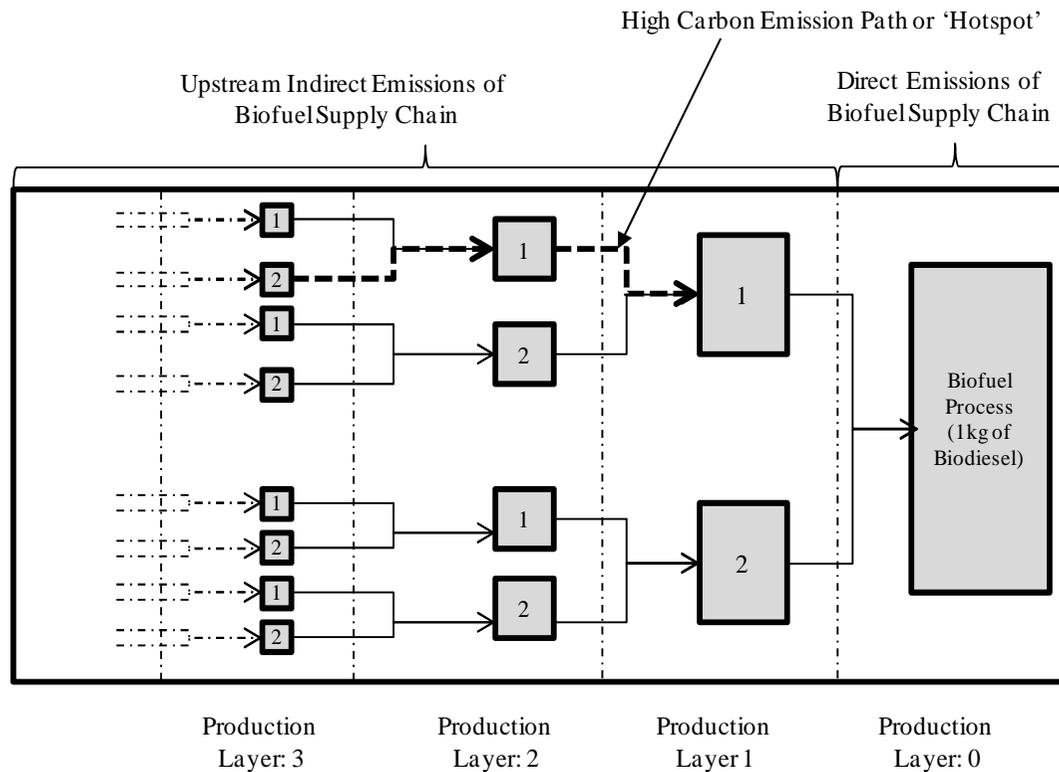
Figure S5: Life Cycle Emissions of RME Production and Supply Chain normalized in energy units
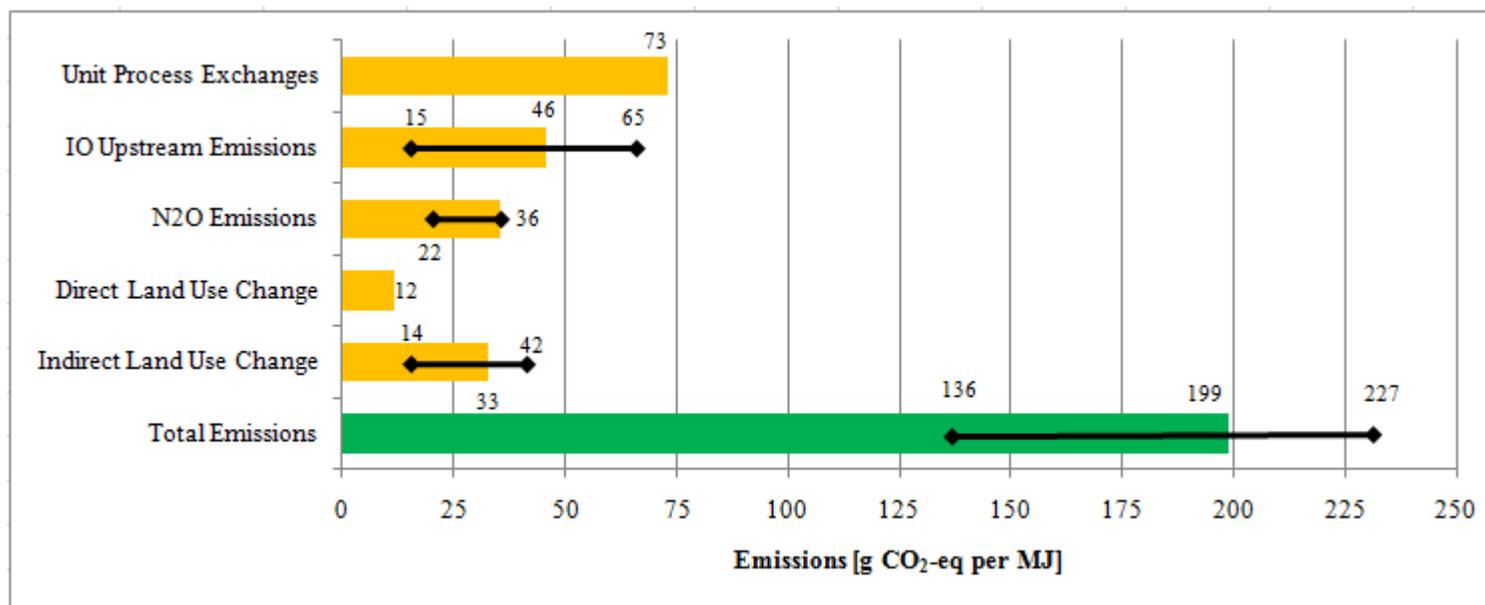
Figure S6: Depiction of build-up of environmental impact along higher upstream production layers in the biodiesel supply chain. The contributions of main sections of the economy are shown.
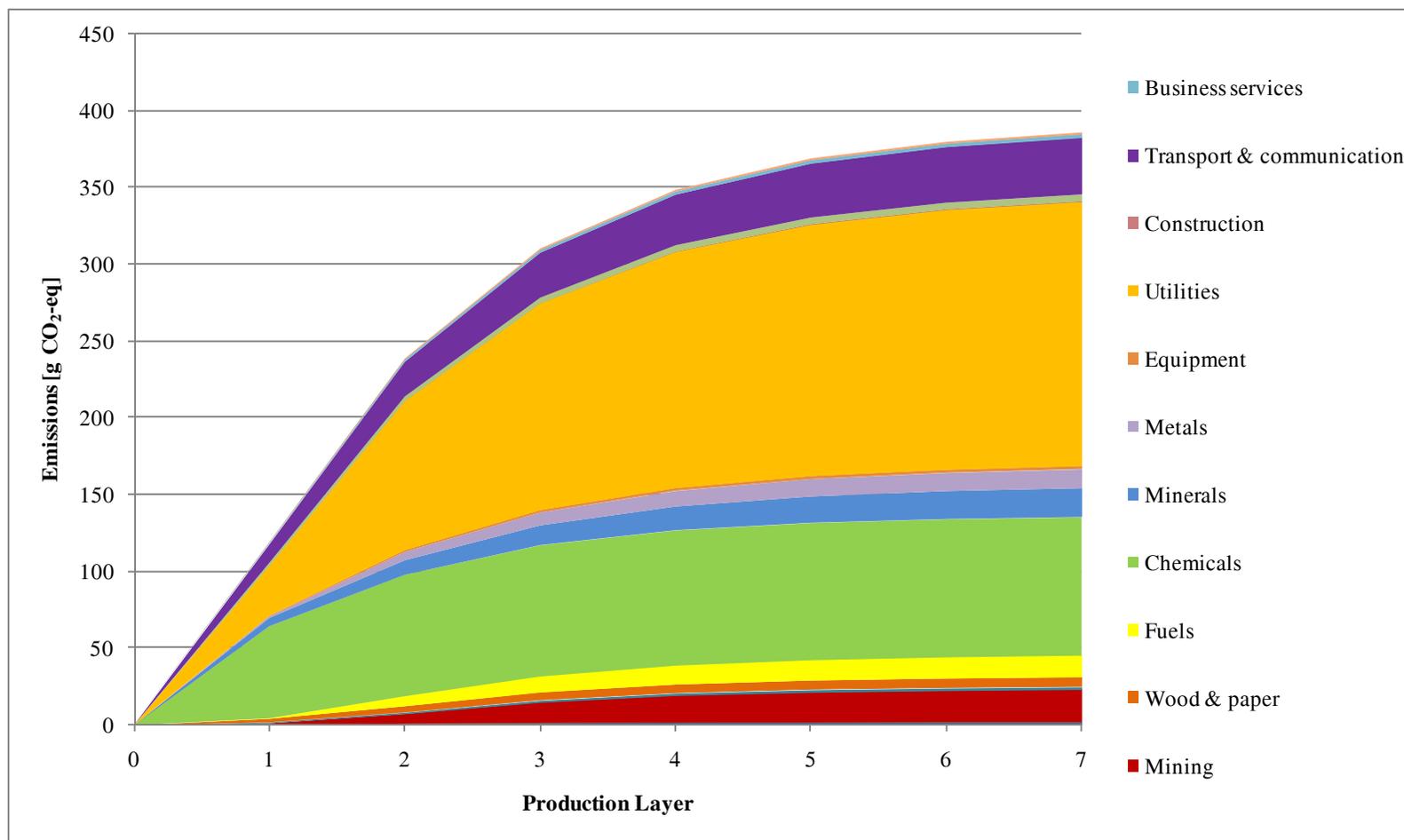
Table S1: Description of Unit Process Exchanges for biodiesel supply chain

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

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

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%

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%