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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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## 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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### 15 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<sub>2</sub>O  
23 emissions from fertiliser application. In this study, a hybrid LCA methodology is used to  
24 evaluate the life cycle CO<sub>2</sub> 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<sub>2</sub>-eq per kg of RME or 36.6% of total life cycle emissions of**  
28 **the RME supply chain.** 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<sub>2</sub>O emissions from fertiliser

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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<sub>3</sub>PO<sub>4</sub>) 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<sub>2</sub>-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

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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

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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<sub>2</sub>O 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

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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:

$$114 \quad \tilde{\mathbf{P}}_{\text{hybrid}} = \begin{bmatrix} \mathbf{E}_p & \mathbf{0} \\ \mathbf{0} & \mathbf{E}_{i-o} \end{bmatrix} \begin{bmatrix} \mathbf{A}_p & -\mathbf{D} \\ -\mathbf{U} & (\mathbf{I} - \mathbf{A}_{i-o}) \end{bmatrix}^{-1} \begin{bmatrix} \mathbf{y} \\ \mathbf{0} \end{bmatrix} \quad \text{(Equation 1)}$$

115 **Where:**

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116  $\tilde{P}_{\text{hybrid}}$  = Total (direct and indirect) environmental impact (e.g. CO<sub>2</sub>-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<sub>2</sub>-eq emissions are  
124 diagonalised (dimension:  $m \times s$ )

125  $E_{i-o}$  = IO environmental extension matrix. CO<sub>2</sub>-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 fromecoinvent 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



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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,  $A_p$  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

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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 fromecoinvent are clearly known:

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178  $P'_{IO} = [E] \times (I - A)^{-1}y = \mathbf{E}Iy + \mathbf{E}A^1y + \mathbf{E}A^2y + \mathbf{E}A^3y + \dots + \mathbf{E}A^ny$  (Equation 2)

179  $EIy'$ , represents the direct GHG emissions emitted (at production level 0) for a given demand  
180 and  $EA^ny$  the indirect GHG emissions emitted for a given final demand at the  $n^{th}$  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  $n^{th}$  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)

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

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.

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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:

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

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<sub>2</sub> 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.

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215

## 216 **2.2 Process Analysis Data**

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

232

## 233 **2.3 Input-Output (IO) Analysis Data**

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

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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<sub>2</sub>-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.**

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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

### 269 **3. Results and Discussions**

#### 270 **3.1 Hybrid Life Cycle CO<sub>2</sub>-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<sub>2</sub>-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<sub>2</sub>-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.

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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<sub>2</sub>O) emissions from the use of fertilisers can potentially be significant enough to  
291 change the environmental profile of biodiesel [35-36].

292 With N<sub>2</sub>O having a global warming potential 298 times that of CO<sub>2</sub> 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<sub>2</sub>O 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<sub>2</sub>O 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



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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<sub>2</sub>O Emissions from Fertiliser Application**

309 The Intergovernmental Panel on Climate Change, IPCC [41] estimate that the direct emission  
310 factor associated with N<sub>2</sub>O 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<sub>2</sub>O emissions had been previously underestimated and  
314 shows that N<sub>2</sub>O emission factor (direct and indirect) in agro-biofuel production is 3–5% of N  
315 applied (that is 0.03-0.05 kg N<sub>2</sub>O-N (kg N)<sup>-1</sup>. 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<sub>2</sub>O to the greenhouse gas balance of first-generation biofuels. The  
320 fertilizer input rate assumed in this study (137.4 kg-N ha<sup>-1</sup> yr<sup>-1</sup>) 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:

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- 323 i. Land Use: Transformation from non-irrigated arable land accounted for 71% of land  
324 use at 2.08 m<sup>2</sup> per kg of rape while transformation from pasture and meadow land  
325 accounted for 29% of land use at 0.85m<sup>2</sup> per kg of rape [45]
- 326 ii. Land Occupation: 11 months per year permanent land-use occupation [46].
- 327 iii. It was deduced fromecoinvent [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<sub>2</sub>-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<sub>2</sub>O emissions factor [kg.N<sub>2</sub>O–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<sup>-1</sup> yr<sup>-1</sup>]

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

337  $k_n$  = Factor to convert from N<sub>2</sub>O–N to N<sub>2</sub>O [equivalent to ( $\frac{44}{28}$ )]

338  $(GWP)_{N_2O}$  = Global Warming Potential of N<sub>2</sub>O.

339  $l$  = Area of land occupied by bioenergy (biodiesel) crop [m<sup>2</sup> 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

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343 Using a GWP of 298 for N<sub>2</sub>O and effective land size of 1.723 m<sup>2</sup> or (1.723 × 10<sup>-4</sup> ha) per  
344 kg of rape seed, it is estimated from Equation 6 that the N<sub>2</sub>O emissions contributed between a  
345 high impact scenario of 1.32 kg CO<sub>2</sub>-eq per kg of RME and a low impact scenario of 0.80 kg  
346 CO<sub>2</sub>-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:

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

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<sub>2</sub>-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

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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<sub>2</sub>-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<sub>2</sub>-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<sub>2</sub>-eq/ha/yr or 1.86 kg CO<sub>2</sub>-eq. Taking into account the  
373 allocation factors in Table 2, iLUC is calculated to be 1.22 kg CO<sub>2</sub>-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<sub>2</sub>-  
378 eq.

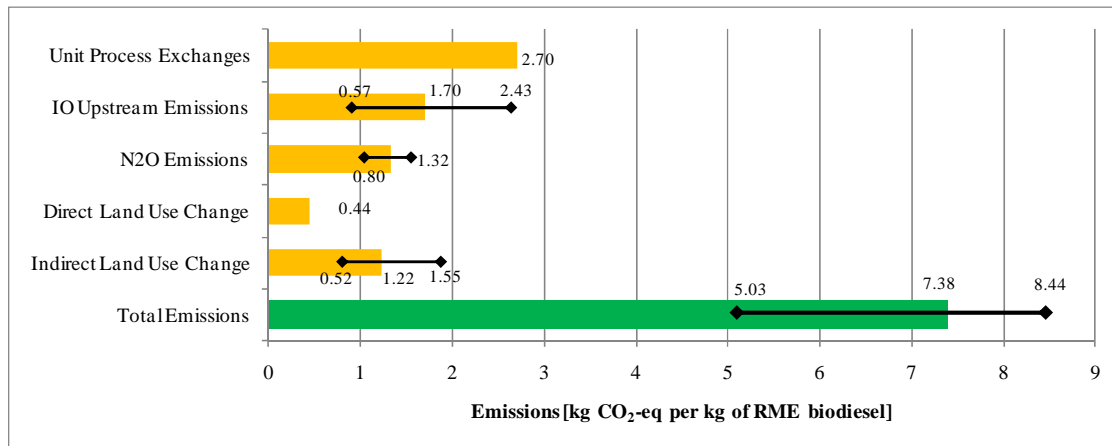
379 The emissions associated with all stages of the RME biodiesel production are shown in  
380 Figure 1. The total life cycle CO<sub>2</sub>-eq emissions for 1kg of RME biodiesel were calculated to  
381 be 7.38 kg CO<sub>2</sub>-eq or 199 g CO<sub>2</sub>-eq/MJ. By accounting for uncertainty in the assessment, it  
382 was estimated that the results are in the range 5.03 to 8.44 kg CO<sub>2</sub>-eq per kg of RME  
383 biodiesel. **Refer to Figure S5 for normalized results in energy units.**

384

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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<sub>2</sub>-eq or 44.5% of total upstream emissions. This was followed by the ‘Chemical Sector’  
394 emitting 90 g CO<sub>2</sub>-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<sub>2</sub>-eq or  
396 9.6%), ‘Mining’ (21 g CO<sub>2</sub>-eq or 5.4%), ‘Minerals’ (19 g CO<sub>2</sub>-eq or 4.8%) and ‘Fuels’  
397 (14 g CO<sub>2</sub>-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<sub>2</sub>-eq of the  
400 high emissions intensity paths or ‘carbon hot-spots’. 150 of the most important paths of the

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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<sub>2</sub>-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<sub>2</sub>-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<sub>2</sub>-eq per kg with an uncertainty range of  
417 5.03 to 8.44 kg CO<sub>2</sub>-eq per kg. The uncertainty was a result of variability in indirect land use  
418 change, N<sub>2</sub>O emissions and IO higher upstream emissions. IO higher upstream emissions  
419 accounted for approximately 23% of total CO<sub>2</sub>-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

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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<sub>2</sub>O  
427 emissions, assuming that the impacts of land use change, N<sub>2</sub>O 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<sub>2</sub>-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<sub>2</sub>O 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<sub>2</sub>O 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<sub>3</sub>PO<sub>4</sub> (85% in water) into the biodiesel**

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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 12<sup>th</sup> 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<sub>2</sub>-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



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472 (CenSA), York, UK (<http://www.bottomline3.co.uk>).

473

474 **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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### 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

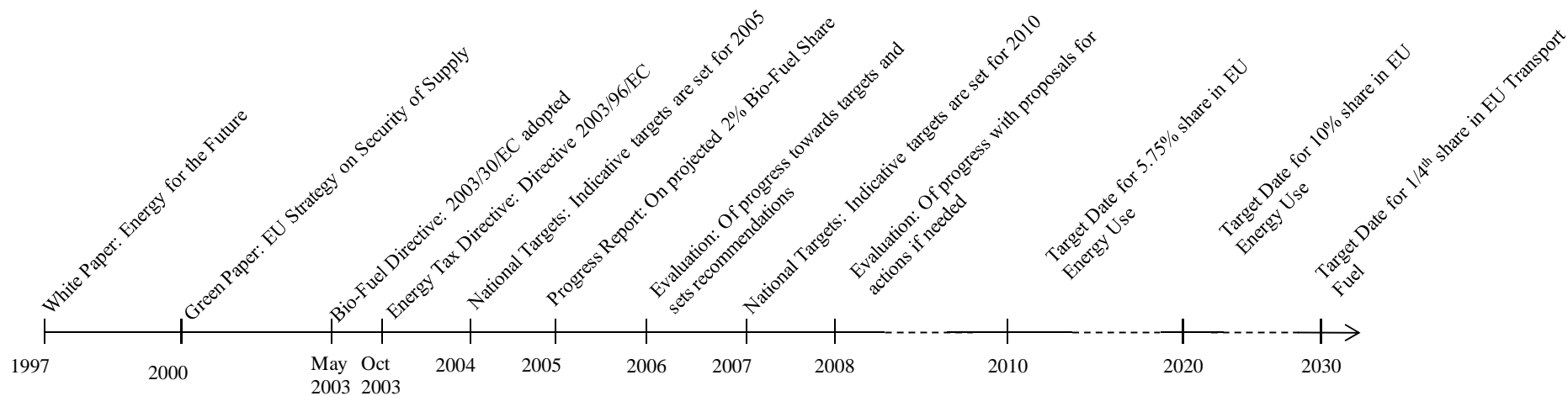
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**Figure S1:** Timeline of Policies affecting Biofuel Production in Europe

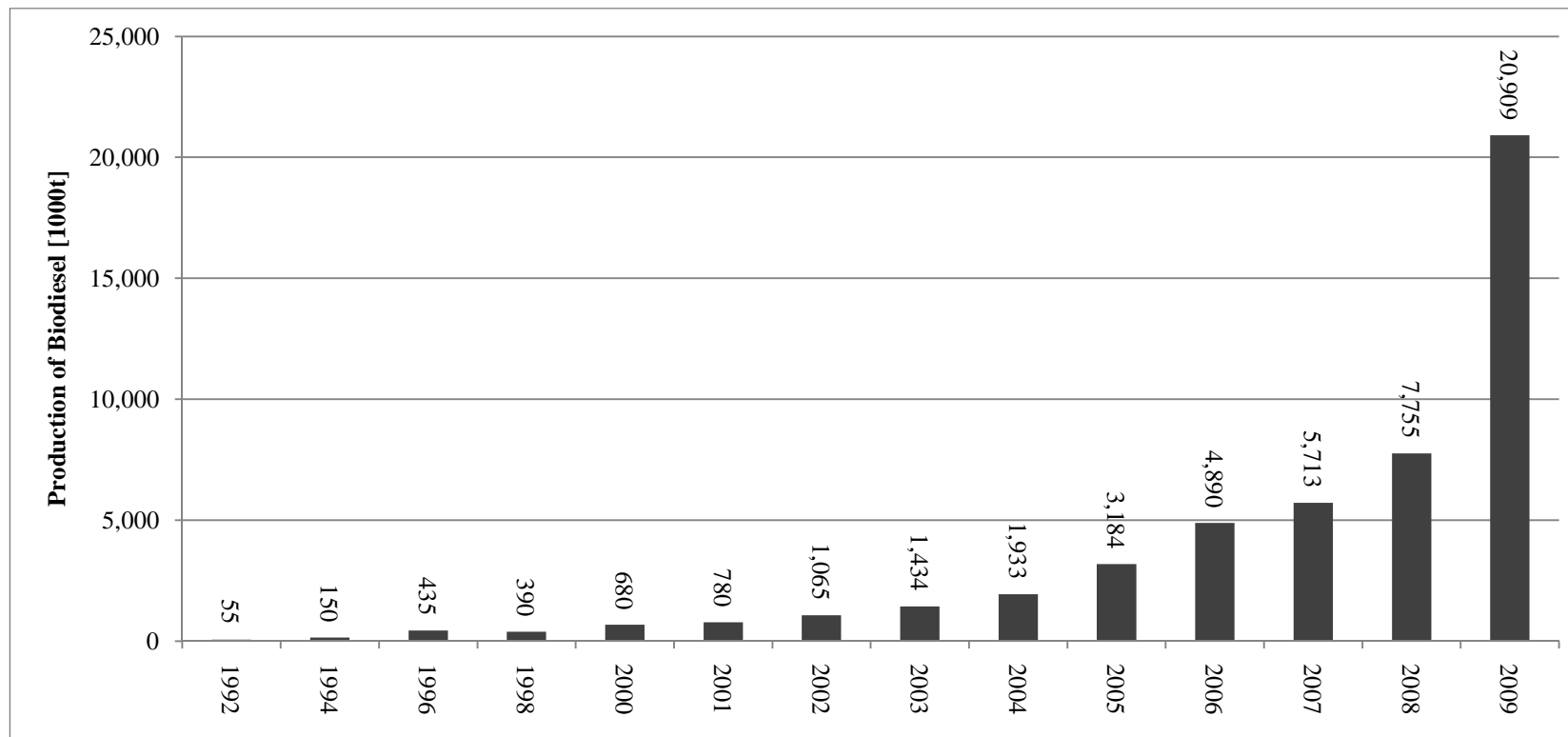


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**Figure S2:** Trend in the growth of Biodiesel in Europe (Adapted from statistics of the European Biodiesel Board, 2009)

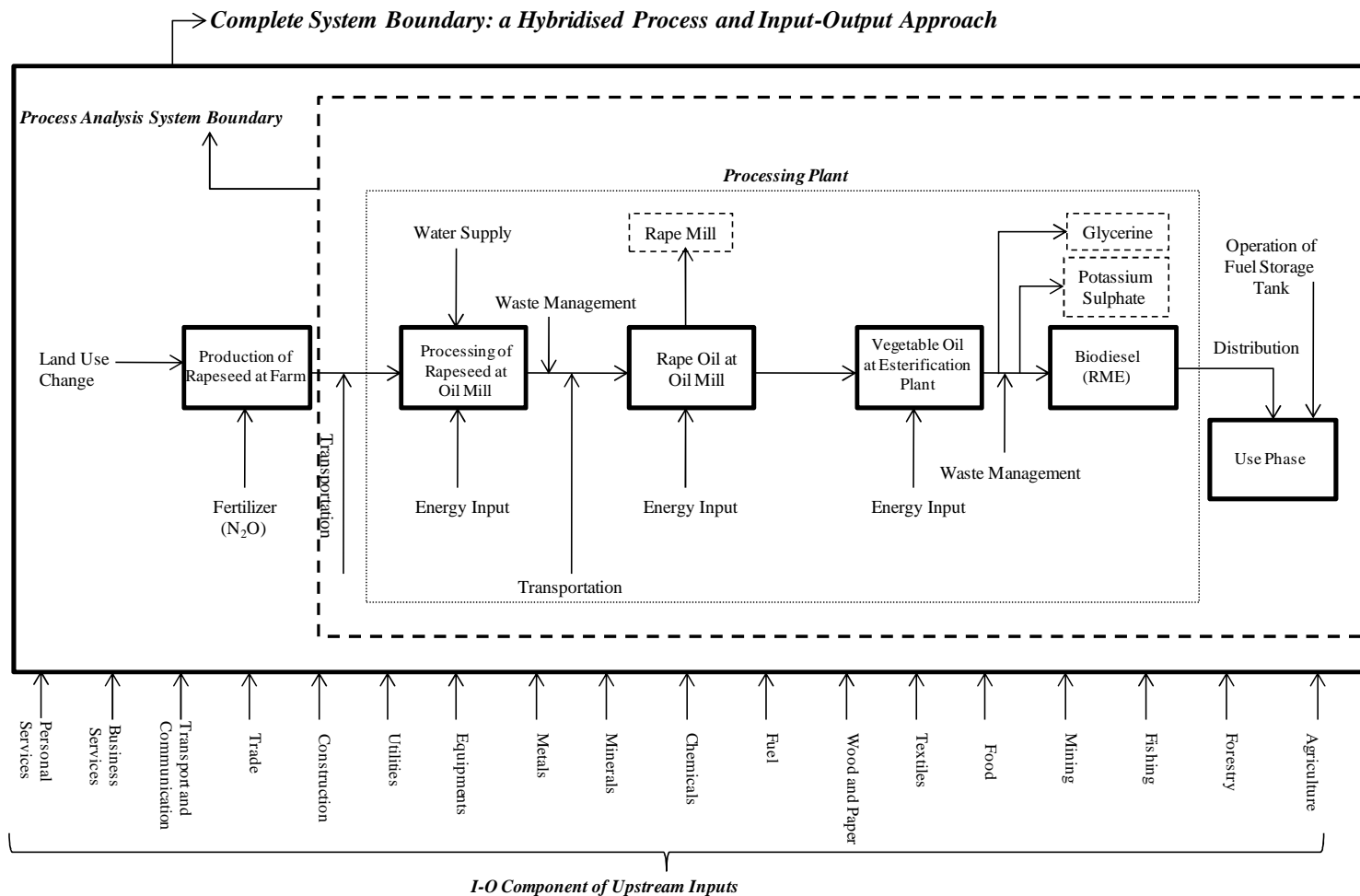


**Reference:** European Biodiesel Board, 2009. "Statistics: The EU Biodiesel Industry". Adapted from: <http://www.ebb-eu.org/stats.php#>

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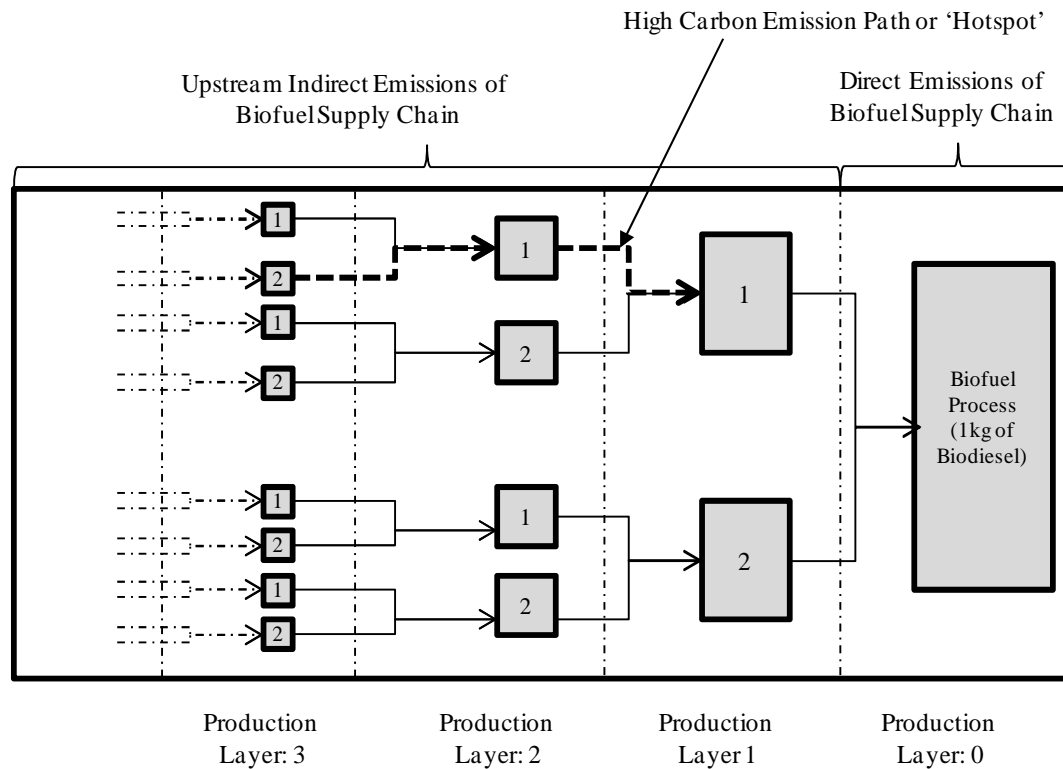
**Figure S3:** Depiction of the Hybridised Systems Approach to GHG Assessment of Biodiesel Supply Chain used in this study



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



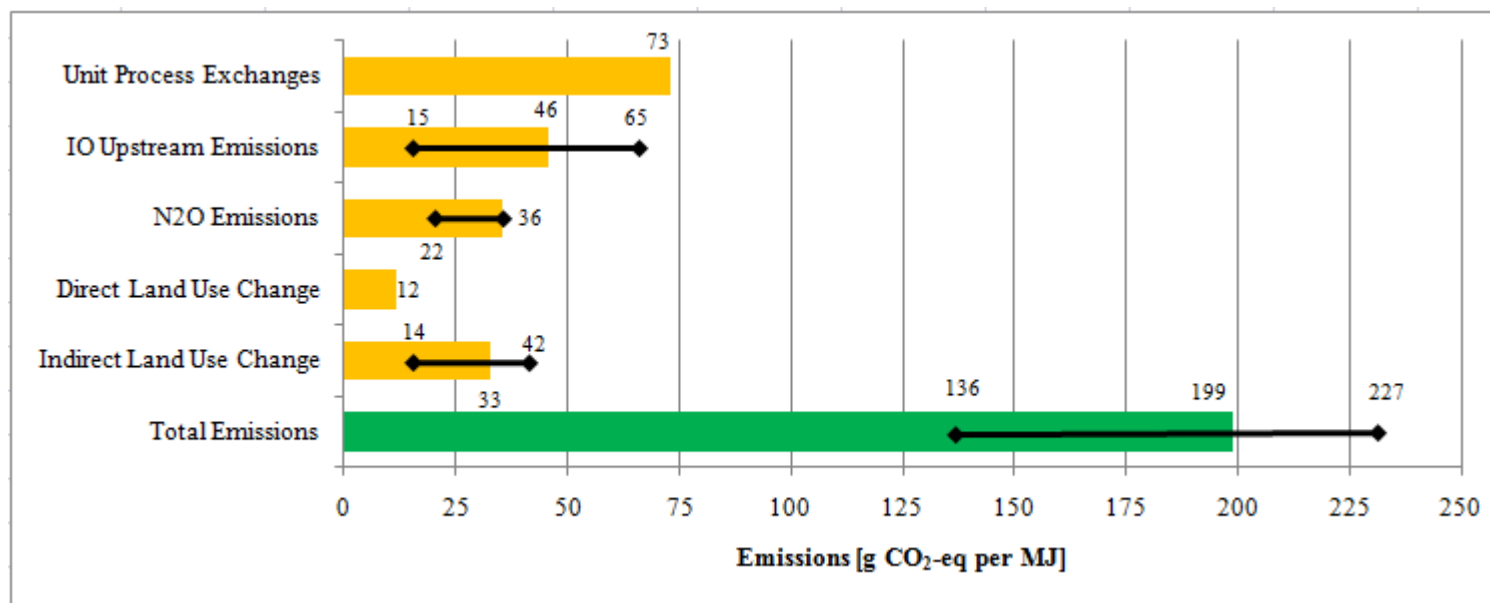


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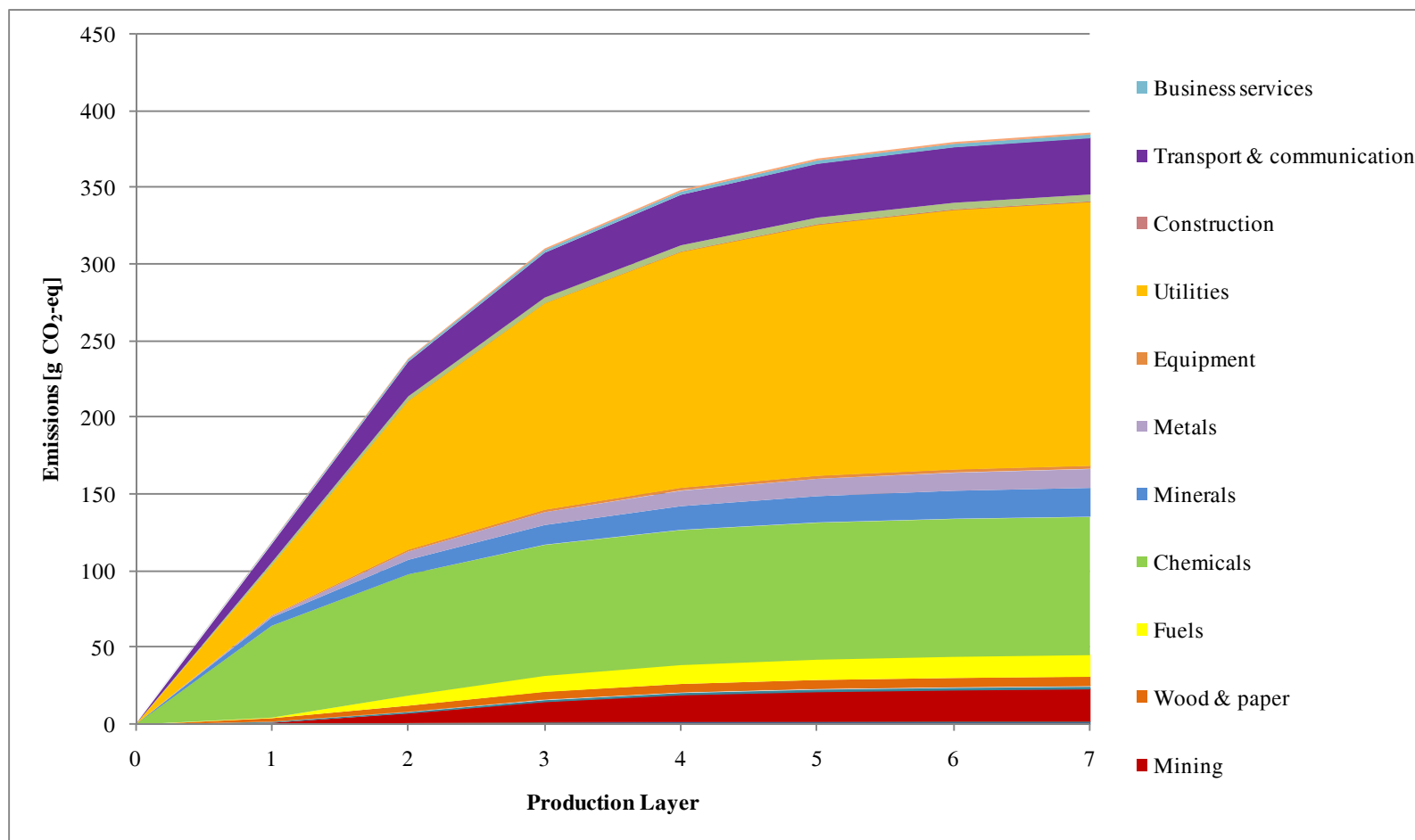
**Figure S5:** Life Cycle Emissions of RME Production and Supply Chain normalized in energy units



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**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

<i>Unit Process Exchanges</i>	<i>Description</i>
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<sup>3</sup> with average storage time of 2 months

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

<i>Production Stage</i>	<i>Products</i>	<i>Allocation Factor (%)</i>
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 <sub>2</sub> -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%

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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 service > 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%