

Future Prospects of Spent Coffee Ground Valorisation Using a Biorefinery

Approach

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Abstract

In the UK, half a million tonnes of spent coffee ground (SCG) waste are generated annually. Current SCG management practices of landfill and energy-from-waste (EfW) facilities either underutilise its valuable constituents or have negative environmental impacts. This study investigates the prospects of SCG-based biorefineries by assessing the impact of biorefinery size, location and products on economic and environmental performances. Two biorefinery design configurations are proposed. Configuration I produces biodiesel and electricity whilst Configuration II produces biodiesel and high-value chemicals. From these configurations, four biorefinery scenarios are analysed at a 10% discount rate with 2019 as the reference year. Configuration I using SCG from London coffee establishments yields a negative net present value (*NPV*) of –£3.9 million and 22% greater greenhouse gas (GHG) emissions than conventional rapeseed biodiesel production. Changing the SCG source to a coffee factory increases biorefinery size by 2.3 times but still produces a negative *NPV*. Using Configuration II to process SCG from coffee shops significantly improves *NPV*. However, without on-site energy generation, its GHG emissions are greater than those from conventional production methods of the high-value chemicals. An on-site Configuration I using SCG from the coffee factory yields the best performance. It produces a *NPV* of £3.1 million and GHG emissions 85% and 13% lower than that of SCG landfilling and conventional biodiesel. Overall, these findings demonstrate the potential of extracting value from SCG waste using a biorefinery approach, revealing a strong likelihood that future SCG biorefineries will be large scale and on-site of SCG production.

27 **Keywords:** Circular economy; spent coffee grounds; biorefinery; biofuels; techno-economic
28 analysis; carbon footprint analysis

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30 **Nomenclature**

AD	Anaerobic digestion
CEPCI	Chemical Engineering Plant Cost Index
EfW	Energy-from-waste
f.o.b	Free on board
FFA	Free fatty acids
GHG	Greenhouse gas
HEN	Heat exchanger network
HMF	Hydroxymethylfurfural
IEA	International Energy Agency
OPEX	Operating expenditure
PHB	Polyhydroxybutyrate
SA	Succinic acid
SCG	Spent coffee ground
CC	Annualised capital cost (£/y)
C_{CARB}	Carbon costs from plant's net CO ₂ produced (£/y)
C_f	Cash flow in a particular year (£)
$COST_b$	Equipment cost at base size and base year (£)
$COST_f$	Equipment free on board cost at base year (£)
C_r	Equipment cost at reference year (£)
CR_E	Emissions credit from electricity exported to grid (kg CO ₂ -eq./t SCG)
CR_F	Capital recovery factor
CR_M	Cost of raw materials (£/y)
CR_P	Emissions credit from displaced use of conventionally produced products (kg CO ₂ -eq./t SCG)
C_U	Cost of utilities (£/y)
C_{WT}	Waste treatment costs (£/y)
dr	Discount rate (%)
E_{COMB}	Emissions from combustion of biodiesel product (kg CO ₂ -eq./t SCG)
E_{ENERGY}	Emissions from energy consumed (kg CO ₂ -eq./t SCG)

<i>EP</i>	Economic potential (£)
<i>E_P</i>	Net direct CO ₂ emissions of plant (kg CO ₂ -eq./t SCG)
<i>E_{RM}</i>	Emissions from raw materials used (kg CO ₂ -eq./t SCG)
<i>E_{TRAN}</i>	Emissions from SCG transport (kg CO ₂ -eq./t SCG)
<i>E_{WT}</i>	Emissions from waste treatment (kg CO ₂ -eq./t SCG)
<i>h</i>	Annual number of operating hours (h)
<i>IV</i>	CEPCI Index value
<i>N_{equip}</i>	Total number of equipment
<i>N_p</i>	Total number of products
<i>NPV</i>	Net present value (£)
<i>OC</i>	Annual operating cost (£/y)
<i>p</i>	Base unit price of product (£/kg)
<i>PL</i>	Plant production lifetime (y)
<i>r</i>	Hourly production rate (kg/h)
<i>R</i>	Scaling exponent
<i>SIZE_b</i>	Equipment base size (units depend on equipment type)
<i>SIZE_f</i>	Equipment current size (units depend on equipment type)
<i>TCC</i>	Total capital cost (£)
<i>VAR</i>	Annual variable operating costs (£/y)
<i>ΔT_{min}</i>	Minimum temperature difference (°C)
<i>Δ%</i>	Percentage change in product price (%)

31

32 **1 Introduction**

33 Coffee is one of the world's most popular beverages. With a global daily consumption of 2.25
34 billion cups, its beans are second only to petroleum as an internationally traded commodity (Giller
35 et al., 2017). Coffee consumption generates large amounts of waste, with the UK alone producing
36 an estimated half a million tonnes of wet spent coffee ground (SCG) annually (Recycling Magazine,
37 2019). Currently, most SCG produced worldwide are disposed in landfills, where its decomposition
38 releases greenhouse gas (GHG) emissions into the atmosphere and acidic leachate containing
39 ecotoxic compounds (Kookos, 2018; Park et al., 2016). SCG is also treated via energy-from-waste
40 (EfW), which allows for energy recovery (Schmidt et al., 2020) but leaves the rich variety of
41 valuable organic compounds found in SCG severely underutilised. Energy recovery from SCG is

42 done by a company known as Bio-bean, which processes the waste into pellets or logs to be sold as
43 renewable solid fuel (Bio-bean, 2021).

44 Insoluble lignocellulosic material (lignin, cellulose and hemicellulose) constitutes almost 70 wt% of
45 dry SCG, while proteins and oils make up 11% and 15%, respectively (Obruca et al., 2015). Most
46 SCG studies considered lignin as a fuel to deliver energy requirements (Mata et al., 2018; Saratale
47 et al., 2020). The only SCG study found to isolate lignin was performed by Lee et al. (2019),
48 utilising the Organosolv pre-treatment. The sugar monomers of SCG cellulose and hemicellulose
49 have been recovered for use in microbial growth and the production of chemicals or materials
50 (Kourmentza et al., 2018). Obruca et al. (2015) and Wang et al. (2010) found that concentrations of
51 fermentable sugars in SCG hydrolysate can be increased through combining dilute acid hydrolysis
52 and enzymatic digestion. Chiyanzu et al. (2015) investigated the application of steam explosion pre-
53 treatment as an effective method to reduce the enzyme dosage required in the ensuing enzymatic
54 hydrolysis of SCG. However, Mayanga-Torres et al. (2017) found that temperature above 150°C
55 rapidly increases the formation of toxic sugar degradation products such as hydroxymethylfurfural
56 (HMF) and furfural, which inhibit microbial activity in subsequent fermentation processes.
57 Kovalcik et al. (2018) suggested that the removal of toxic co-contaminants present in SCG
58 hydrolysates via detoxification steps may increase utilisation of the waste. Using different
59 microorganism strains, the fermentation of recovered SCG sugars can yield a wide range of
60 products, such as bioethanol, bioplastics and high-value platform chemicals (Kourmentza et al.,
61 2018).

62 On average, SCG contains around 15% oil (Kondamudi et al., 2008). Synthesis of biodiesel from
63 SCG oil via transesterification has been demonstrated (Al-Hamamre et al., 2012; Kondamudi et al.,
64 2008), where transesterification is the reaction between triglycerides and lower alcohols. These
65 studies used solvent extraction methods to isolate the oil from SCG. To eliminate the need for a
66 separate oil extraction process, the *in-situ* transesterification of SCG has been investigated, where
67 simultaneous extraction and transesterification produced biodiesel (Tuntiwiwattanapun et al., 2017).

68 However, this procedure has been criticised for its high energy consumption (Kookos, 2018). One
69 of the challenges of SCG oil transesterification arises due to its high free fatty acid (FFA) content,
70 which reacts with alkali catalysts to form soap (Tuntiwiwattanapun et al., 2017). To avoid
71 significant soap formation and the deactivation of the alkaline catalyst in transesterification, the
72 SCG must be of less than 3 wt% FFA (Trejo-Zárraga et al., 2018). To address this, Caetano et al.
73 (2014) performed acid esterification and alkaline transesterification on SCG oil, resulting in a 37%
74 increase in biodiesel yields when compared against the direct transesterification of SCG. Typical
75 yields of biodiesel from esterification and transesterification have been found to be around 0.84 kg
76 biodiesel/kg oil (Abdelaziz et al., 2020, 2019). Energy recovery from SCG has been explored
77 extensively in literature. Various studies showed that dried SCG has a lower heating value (18.8
78 MJ/kg at 10% moisture) greater than or comparable to conventional biomass (16.3-17.6 MJ/kg for
79 wood pellet with 7-10% moisture) but lower than fossil fuels (25-42.5 MJ/kg), and thus it is suitable
80 to be used as a fuel in direct combustion (Kang et al., 2017; McNutt and He, 2018; Mayson and
81 Williams, 2021). However, using pure SCG as boiler fuel may cause unstable combustion, leading
82 to lower boiler efficiency and elevated levels of toxic gas and particle emissions (Allesina et al.,
83 2017; Kang et al., 2017). These issues can be minimised by blending SCG with sawdust, resulting
84 in combustion parameters similar to that of wood pellets (Limousy et al., 2012). Li et al. (2014),
85 Kelkar et al. (2015), and Choi et al. (2017) investigated SCG valorisation via fast pyrolysis, which
86 involves the thermochemical decomposition of biomass into bio-oil, char, and non-condensable gas
87 at high temperatures and in an oxygen-free atmosphere. These studies demonstrated high SCG
88 conversion, with Kelkar et al. (2015) reporting SCG conversion rates of 60%, 17.7% and 20.4% for
89 bio-oil, char and pyrolysis gas when treated at 500°C and 32 seconds. A recent study by Kibret et al.
90 (2021) achieved a 95% carbon conversion at 900°C via gasification of SCG in steam and CO₂, a
91 process which also involves biomass decomposition under high temperatures. Besides that, studies
92 on the anaerobic bacterial decomposition of SCG to methane rich biogas have been performed since
93 the 1980s. This process is known as anaerobic digestion (AD). Lane (1983) and Dinsdale et al.

94 (1996) found SCG to have limited application as a mono-substrate of AD, failing to achieve stable
95 biogas production due to inhibitory compounds. To circumvent this, studies such as that by Kim et
96 al. (2016) have investigated the co-digestion of SCG with various co-substrates. As environmental
97 and resource security issues continue to rise, there has been increasing policy focus on the adoption
98 of a circular economy. Circular economy concept aims to promote reduction of resource
99 consumption and waste to landfills through reusing, recycling or converting waste materials into
100 more useful products (Department for Environment, Food and Rural Affairs (DEFRA), the
101 Department of Agriculture, Environment and Rural Affairs (DAERA), the Welsh Government and
102 the Scottish Government, 2020; Ng et al., 2019). The International Energy Agency (IEA)
103 highlighted the importance of implementing a biorefinery approach (i.e. facility with integrated
104 processes that convert biomass into value-added products) to achieve higher value generation and
105 sustainability (Ng et al., 2017; van Ree et al., 2012). It also identified the main driver of biorefinery
106 development as the efficient, cost-effective production of transportation biofuels. Furthermore, the
107 IEA proposed the production of high-value products alongside biofuel products as a promising
108 approach to reduce biofuel production costs within biorefineries (Sadhukhan et al., 2014).

109 SCG has been found to have great potential as a feedstock for the production of a wide range of
110 chemicals and materials, including biofuels (Massaya et al., 2019). However, few studies have
111 investigated the techno-economic performance of SCG biorefineries. Giller et al. (2017), Kookos
112 (2018) and Thoppil and Zein (2021) performed economic evaluations on small-scale biodiesel
113 production plants without the incorporation of high-value products. Kookos (2018) highlighted the
114 importance of large plant scales to achieve competitively priced biodiesel from SCG. The study also
115 mentioned that large scale would involve economical and practical difficulties, such as increased
116 logistical challenges and costs for SCG collection. Thoppil and Zein (2021) emphasised that
117 sensitivity analyses using different product price scenarios are needed to rigorously assess the
118 viability of producing biodiesel from SCG. There is thus a lack of literature evaluating the
119 economic and environmental performance of SCG biorefineries. These assessments are vital for

120 informing the commercial viability of SCG biorefineries and thus their future prospects (Karmee,
121 2018).

122 This study addresses the gap in literature by investigating how the economic and environmental
123 performance of SCG biorefineries are affected by the following key factors: (a) plant size; (b)
124 valorisation routes; (c) plant location relative to SCG source. Factors (a) and (b) have been selected
125 due to their significant influence on plant economics (Kookos, 2018; van Ree et al., 2012), whilst
126 (c) has been chosen to assess biorefinery economics when SCG collection costs of a large
127 biorefinery is removed by locating the plant within an instant coffee factory site. Four biorefinery
128 scenarios of different scale, valorisation routes, and locations have been analysed. The process
129 configurations focus on biodiesel production as transportation biofuels are the main drivers of
130 biorefinery development and there is a substantial amount of literature material available regarding
131 SCG valorisation to biodiesel.

132 The performances of these biorefinery systems have been compared against that of conventional
133 SCG management practices and biodiesel production processes. Additionally, the GHG emissions
134 and costs of the biorefineries have been analysed to help identify what future research efforts should
135 focus on to improve the economic and environmental performance of SCG biorefineries. The
136 novelty of this research lies in the creation of innovative conceptual biorefinery designs for SCG
137 valorisation that incorporate resource efficiency enhancement features such as multiple product
138 generation and energy integration, followed by detailed techno-economic and environmental impact
139 evaluation on different scenarios that consider plant size, valorisation routes and plant location. As
140 environmental pressure from food waste and urgency to decarbonise transport fuels escalates, this
141 research is timely and crucial to identify the most economically and environmentally compelling
142 strategies to both tackle SCG and provide society with low-carbon advanced biofuel.

143 2 Methodology

144 Literature review was performed to gain a holistic view of the SCG valorisation routes explored in
145 previous studies. The market values of the products from these routes, chemical composition of
146 SCG and its availability in the UK were gathered. Two biorefinery configurations were proposed.
147 As the transportation sector is a crucial driver of biorefinery development, both configurations
148 aimed to produce biodiesel. Configuration I was based on a well-researched SCG valorisation
149 pathway (i.e., transesterification) combined with current practices (i.e., incineration with energy
150 recovery). Configuration II also aimed to maximise value extracted by incorporating more novel
151 valorisation methods for the production of high-value chemicals. Four biorefinery scenarios were
152 generated, with different combinations of biorefinery size, design configuration, and location
153 relative to the SCG feedstock source. For each scenario, process simulation modelling and heat
154 integration were performed. Technical, economic, and environmental performances were then
155 evaluated using energy and mass flow data from the simulations. The key indicators used to
156 compare the scenarios include biodiesel yield, economic potential, net present value, cost of
157 production per litre of biodiesel and greenhouse gas emissions.

158 **Process Simulation Modelling:** Aspen Plus V11 software was used to simulate the mass and
159 energy flows of each system. Overall process flow diagrams were developed, with operating
160 conditions and reaction pathways established based on experimental results from literature. The
161 non-random two-liquid (NRTL) thermodynamic property method was used as non-ideal solutions
162 were simulated (Gómez-Ríos et al., 2019). Distillation columns were simulated via Aspen RadFrac
163 models with partial-vapour condensers. Pinch analysis was used to analyse the process heat
164 recovery potential and establish targets for minimum utility consumption. The mass flowrate,
165 enthalpy change, initial and final temperatures of hot and cold streams were extracted from the
166 Aspen Plus simulation results and input into Aspen Energy Analyser V11 software, along with a
167 specified global minimum temperature difference (ΔT_{min}). Using this data, a heat exchanger network
168 (HEN) design was generated and its heat exchangers used in capital and operating cost evaluations.

169 **Economic assessment:** The economic analysis involved the evaluation of capital expenditure,
 170 operating expenditure (OPEX), and profitability. An exchange rate of 1GBP = 0.72 USD was
 171 applied. The total installed capital cost (*TCC*) of a plant was categorised into direct and indirect
 172 costs. The direct costs included the free-on-board (f.o.b.) purchase and installation costs of the
 173 biorefinery equipment. Indirect capital costs included costs for site engineering, designing, and
 174 building. To estimate *TCC*, the Lang's method was used. Equipment f.o.b cost was calculated and
 175 updated to the reference year using the CEPCI index. This is shown in Equations (1)–(2), where
 176 $COST_b$ and $COST_f$ are the equipment base and purchase costs; $SIZE_b$ and $SIZE_c$ are the size capacities of
 177 the base and current systems; R is the scaling exponent; C_r is the cost; IV_r and IV_b the index values
 178 of the reference year (2019) and base year (Davis et al., 2013). Finally, *TCC* was calculated using
 179 Equation (3), where N_{equip} is the total number of equipment and 5.03 is the Lang factor of a solid-
 180 fluid processing biorefinery (Sadhukhan et al., 2014). This method of estimating capital investment
 181 gives a 30% accuracy. However, it must be noted that the investment required for a new plant on an
 182 undeveloped site will be larger than on an existing, developed site. The annualised capital cost (*CC*)
 183 was obtained by spreading *TCC* over the plant's lifetime (*PL*) of 20 years at a discount rate (*dr*) of
 184 10%, which is similar to the discount rates used in previous biorefinery economic analyses
 185 (Zetterholm et al., 2020). *CC* was estimated by using a capital recovery factor (*CRF*) as in
 186 Equations (4)–(5) (Ng and Martinez-Hernandez, 2020).

$$\frac{COST_f}{COST_b} = \left(\frac{SIZE_c}{SIZE_b} \right)^R \quad (1)$$

$$C_r = COST_f \left(\frac{IV_r}{IV_b} \right) \quad (2)$$

$$TCC = 5.03 \times \sum_{i=1}^{n=N_{equip}} C_{r,i} \quad (3)$$

$$CRF = \frac{dr(1+dr)^{PL}}{(1+dr)^{PL}-1} \quad (4)$$

$$CC = TCC \times CRF \quad (5)$$

187

188 OPEX included fixed and variable operating costs. Fixed operating cost comprised of the costs of
 189 labour, supervision, laboratory, plant overhead, maintenance, local taxes and insurance (see
 190 Appendix B, Table B.1). The number of personnel needed per shift was calculated by multiplying
 191 the number of plant processing steps with the number of personnel required per step. The annual
 192 cost of labour was calculated using Equation (6). The annual variable operating cost of the plant
 193 (VAR) [£/y] was calculated using Equation (7), where C_{RM} [£/y] represents raw material cost, C_U
 194 [£/y] the utilities cost, C_{WT} [£/y] the waste treatment costs and C_{CARB} [£/y] the carbon costs based on
 195 the plant's net flowrate of CO₂. Economic viability was measured using key performance
 196 indicators such the economic potential (EP) [£], net present value (NPV) [£] and payback period.
 197 EP and NPV were calculated using Equations (8)–(9), where h [h] is the number of operating hours
 198 annually; r_i [kg/h] and p_i [£/kg] are the production rate and base unit price of product i respectively;
 199 N_p is the total number of products; OC [£/y] is the annual operating costs and C_f [£] refers to the
 200 cash flow in a particular year (Ng et al., 2013).

$$\text{Cost of personnel (\pounds/year)} = \text{Number of personnel per shift} \times 5 \text{ shifts} \times 40 \text{ hours/week} \times \quad (6)$$

$$52 \text{ weeks/year} \times \text{Hourly wage}$$

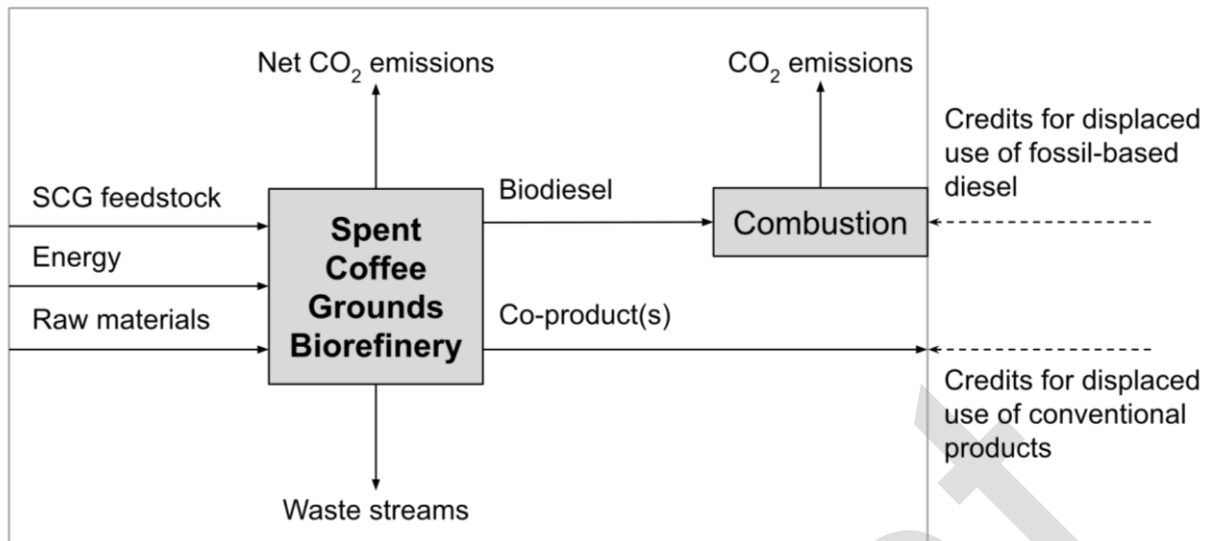
$$VAR = C_{RM} + C_U + C_{WT} + C_{CARB} \quad (7)$$

$$EP = h \sum_{i=1}^{i=N_p} r_i p_i - CC - OC \quad (8)$$

$$NPV = \sum_{n=0}^{n=PL} \frac{C_f}{(1+dr)^n} \quad (9)$$

201

202 **Environmental impact assessment:** The system boundary was defined (see Figure 1) and
 203 inventory analysis was performed.



204

205 Figure 1. System boundary for the SCG biorefinery.

206 Input and output stream data were compiled from simulation results. CCaLC2 (a life cycle
 207 assessment software developed by the University of Manchester) was used to estimate the GHG
 208 emissions of these input and output flows. Using ISO 14067 standards (International Organization
 209 for Standardization, 2018), the net GHG emissions of the SCG biorefinery was calculated using
 210 Equation (10), where E_{RM} , E_P , E_{COMB} , E_{ENERGY} , E_{TRAN} , E_{WT} are the emissions from the raw materials
 211 used, the plant processes, the combustion of the biodiesel produced, the energy consumed, SCG
 212 transport, and waste treatment. Emission credits (GHG removals) for the electricity exported to the
 213 grid (CR_E) and displacement of conventional production of the products generated from SCG (CR_P)
 214 are applied.

$$\text{Net GHG} = E_{RM} + E_P + E_{COMB} + E_{ENERGY} + E_{TRAN} + E_{WT} - CR_E - CR_P \quad (10)$$

215 **Sensitivity analysis:** Price sensitivity analysis was performed on all scenarios using methodology
 216 outlined by Ng et al. (2013). A percentage change in price ($\Delta\%$) was defined and three sets of prices
 217 were generated for each product: the base price (subscript 0), a price $\Delta\%$ lower than the base price
 218 (subscript $-\Delta\%$), and a price $\Delta\%$ higher than the base price (subscript $+\Delta\%$). For N_p number of
 219 products, there are n possible combinations of prices. MATLAB was used to calculate the revenues
 220 of each of these combinations using the equation shown in Equation (11), where the price scenarios
 221 matrix was multiplied by production rates (r) to give n revenue scenarios. The capital and operating

222 costs (assumed to be fixed) were subtracted from each element of the revenue column vector
 223 produced in Equation (11) to give the *EP* for each price scenario.

224 An economic risk is a change in revenue due to change in the combination of product prices.
 225 Classes of economic risks were defined in terms of the percentage change in *EP* relative to the *EP*
 226 calculated from base prices. The probability of a class of economic risk occurring is the ratio of the
 227 number of combinations that produce that class of economic risk to the total number of
 228 combinations.

$$\begin{bmatrix} p_{1,0} & p_{2,0} & \cdots & p_{Np,0} \\ p_{1,+\Delta\%} & p_{2,+\Delta\%} & \cdots & p_{Np,+\Delta\%} \\ p_{1,-\Delta\%} & p_{2,-\Delta\%} & \cdots & p_{Np,-\Delta\%} \\ \vdots & \vdots & \vdots & \vdots \end{bmatrix}_{n \times Np} \begin{bmatrix} r_1 \\ r_2 \\ \vdots \\ r_{Np} \end{bmatrix}_{Np \times 1} = \begin{bmatrix} p_{1,0} r_1 + \cdots + p_{Np,0} r_{Np} \\ p_{1,+\Delta\%} r_1 + \cdots + p_{Np,+\Delta\%} r_{Np} \\ p_{1,-\Delta\%} r_1 + \cdots + p_{Np,-\Delta\%} r_{Np} \\ \vdots \end{bmatrix}_{n \times 1} \quad (11)$$

229

230

231 3 SCG Biorefinery Configurations: Modelling and Integration

232 This section details the two biorefinery configurations proposed. Process description and mass and
 233 energy balances are presented. Detailed stream tables, process flowsheets and the specifications
 234 used in Aspen Plus process models are provided in Appendix C.

235 3.1 Proposed SCG Configurations

236 In the biorefinery configuration descriptions, a wet SCG plant input of flowrate 5767 kg/h at 20°C
 237 was employed based on SCG produced by London coffee establishments (see Section 3.2).
 238 Configuration I produces biodiesel from extracted SCG oil, with electricity generated as a co-
 239 product due to the combustion of defatted SCG. Heat from the combustion is exported to a heat
 240 network. Configuration II produces biodiesel as well as succinic acid (SA), aromatics (vanillin,
 241 vanillic acid, guaiacol, acetovanillone), carboxylic acids (formic acid, acetic acid) and
 242 polyhydroxybutyrate (PHB) bioplastic. In contrast to Configuration I, no combustion is performed.
 243 Heat integration in both configurations uses a ΔT_{min} of 15°C.

244 3.2 SCG Feedstock

245 **SCG feedstock composition:** SCG feedstock was modelled with a moisture content of 66%
246 (Caetano et al., 2014). Dry mass compositional data and their representative Aspen Plus
247 components are presented in Table 1 (Obruca et al., 2015). Further details about the component data
248 used in the simulations can be found in Table A.1 of Appendix A.

249 Table 1. Chemical composition of SCG dry mass and the representative components adopted in
250 Aspen Plus (Obruca et al., 2015).

SCG Component	Representative component adopted in Aspen Plus	Content⁽ⁱ⁾ (wt%)
Cellulose	Cellulose	10.95
Hemicellulose	Cellulose	31.83
Lignin	Vanillin	27.00
Oil (triglycerides)	Triolein	11.35
Oil (free fatty acids)	Oleic acid	3.65
Protein ^(iv)	Glutamic acid ⁽ⁱⁱ⁾	10.70
Polyphenol	Caffeic acid ⁽ⁱⁱⁱ⁾	2.50
Caffeine	Caffeine	0.02
Ash ^(iv)	Calcium Oxide	2.00
Moisture	Water	66

251

252 Note:

253 ⁽ⁱ⁾ All components are in wt% of dry mass except for water.

254 ⁽ⁱⁱ⁾ Glutamic acid found to be major component of SCG protein (Martinez-Saez et al., 2017).

255 ⁽ⁱⁱⁱ⁾ Caffeic acid identified in SCG (Panusa et al., 2013).

256 ^(iv) Protein and ash treated as insoluble compounds and did not partake in any chemical reaction (Humbird et al., 2011).

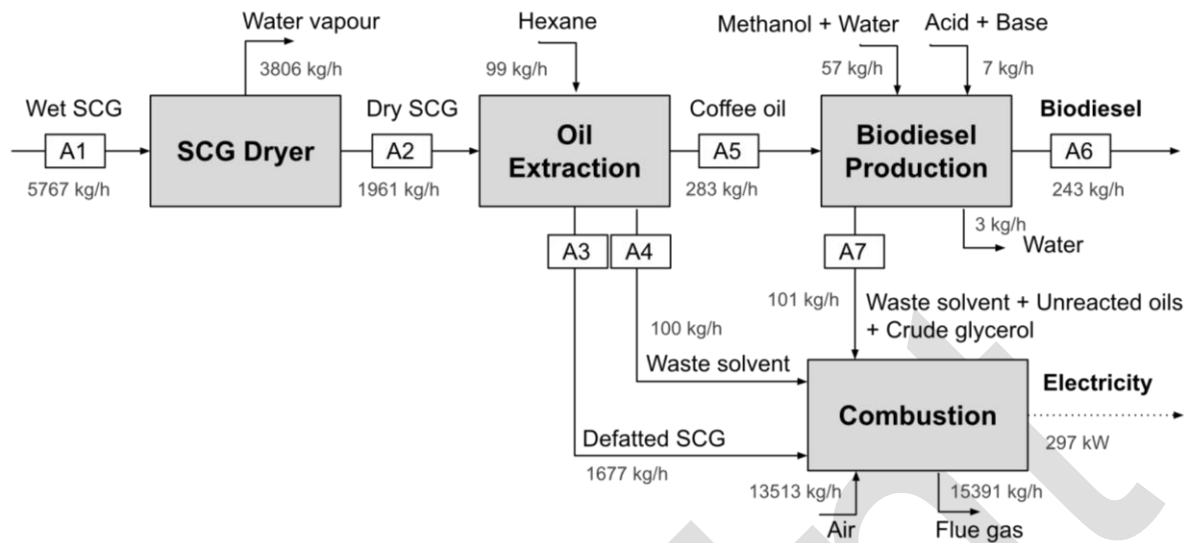
257 **SCG feedstock availability:** The SCG used in the scenarios was obtained from either London
258 coffee establishments or Nestlé's coffee factory in South Derbyshire, UK. The parameters used to
259 calculate the amount of SCG produced in London coffee shops are summarised in Table A.3 of
260 Appendix A. The total amount of wet SCG produced in London coffee establishments was
261 calculated using Equation (A.1), giving a value of 46,136 t/y. On the other hand, the annual wet
262 SCG production rate from Nestlé's coffee factory was calculated as 105882 t/y using Equation
263 (A.2) and the parameters in Table A.4.

264 **3.3 Biorefinery Scenarios**

265 This section outlines four SCG biorefinery scenarios based on Configurations I and II, described in
266 Sections 3.5 and 3.5. The biorefinery plants in Scenarios A, B and D were taken to be 70 km away
267 from their SCG source, based on the average distance between the centre of Greater London and its
268 perimeter. Other parameters used for SCG transport are included in Table A.2 in Appendix A.
269 Besides this, input SCG feedstock of these 3 scenarios enters the biorefineries at 20°C. Scenario A
270 is considered the base case scenario. Here, Configuration I processes all the SCG generated in
271 London coffee shops, producing biodiesel and electricity. The parameters used in Scenario B are
272 identical to those of Scenario A except for the use of all the SCG produced from the Nestlé coffee
273 factory. On the other hand, Scenario C assumes the biorefinery of Scenario B is located within the
274 coffee factory site and that the SCG is delivered directly from the factory's coffee brewing process,
275 entering the biorefinery at 80°C. SCG transportation is excluded in Scenario C and the SCG
276 feedstock is thus obtained at no cost. Scenario D uses parameters identical to that of Scenario A
277 apart from the use of Configuration II.

278 **3.4 Configuration I**

279 The flow diagram is illustrated in Figure 2.



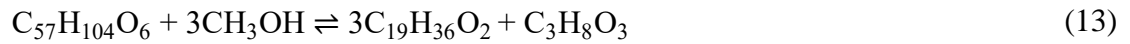
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282 Figure 2. Flow diagram of SCG biorefinery Configuration I.

283 **SCG drying and oil extraction:** Wet SCG (stream A1) is dried at 102°C to reduce moisture
 284 content from 66% to 0%. As moisture reduces oil solubility in the hydrophobic solvent, the absence
 285 of water is crucial to achieve high oil extraction yields in the following process (Najdanovic-Visak
 286 et al., 2017). Solvent extraction of the SCG oil content (FFAs and triglycerides) is performed on dry
 287 SCG (stream A2) at 60°C, with process parameters summarised in Table C.1 of Appendix C
 288 (Najdanovic-Visak et al., 2017). *n*-hexane is used as it has been found to give the highest yield of
 289 oil (Somnuk et al., 2017). The liquid solvent/oil mixture and the solid defatted SCG are separated
 290 via centrifugation and the solvent is removed in a flash vessel at 0.1 bar and 80°C. The separated
 291 solvent is recycled for further use in oil extraction. The defatted SCG (stream A3) and waste solvent
 292 stream from the recycle loop purge (stream A4) are combusted whilst the extracted coffee oil
 293 (stream A5) is used in biodiesel production.

294 **Biodiesel production from SCG oil:** Oil (stream A5) enters a two-step process to produce
 295 biodiesel (Efthymiopoulos et al., 2018; Hochegger et al., 2019; Saratale et al., 2020). Esterification
 296 is performed before transesterification to esterify the FFA and minimise soap formation. Table C.2
 297 summarises the process parameters used. During the esterification process described in Equation
 298 (12), methanol (CH₃OH) reacts with FFA (C₁₈H₃₄O₂) to form biodiesel (C₁₉H₃₆O₂) with sulphuric

299 acid as the catalyst. The products of esterification undergo sodium hydroxide (NaOH) catalysed
300 transesterification where biodiesel and glycerol (C₃H₈O₃) are formed, as shown in Equation (13).



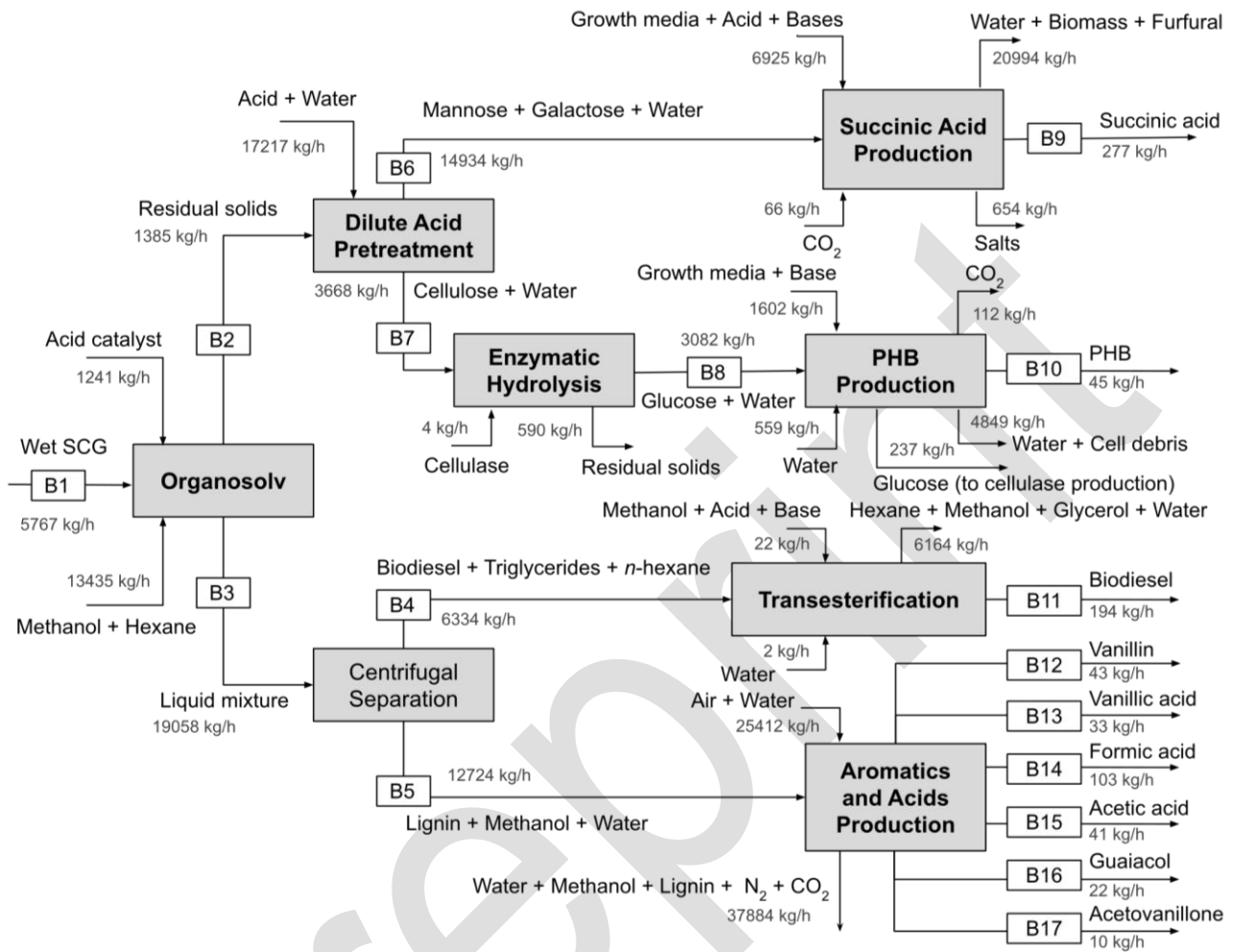
301 The transesterification products enter a flash vessel at 162°C and 1 bar to recover methanol for
302 recycling. The bottom products of the flash vessel are washed in acidic water to neutralize the base
303 catalyst used in transesterification. Next, centrifugation separates biodiesel, methanol, and hexane
304 from the other denser liquids. Two flash vessels at 0.1 bar and 167°C and 270°C purify the
305 biodiesel, producing biodiesel (stream A6) to EN14214 European standards (Tsoutsos et al., 2019).
306 The crude glycerol produced from transesterification, methanol recycle-loop purge and other waste
307 organic streams consisting of unreacted oils (stream A7) are combusted.

308 **Combustion of defatted SCG and waste streams:** The combustion of streams A3, A4 and A7 was
309 modelled using a stoichiometric reactor at 500°C and 1 bar. Flue gas exiting the combustion
310 chamber at 500°C is used to pre-heat boiler feed water from 20°C to 102°C, reducing the gas
311 temperature to 112°C. The heat from combustion reactor is used to generate steam of 480°C and
312 135 bar from the pre-heated water. The steam drives a turbine of 90% isentropic efficiency and a
313 pressure drop of 107.5 bar, generating a total of 1085 kW of electricity. Of this, 788 kW is used by
314 plant processes and the remaining 297 kW is exported to the grid. Heat from the steam leaving the
315 turbine (247°C and 27.5 bar) and the flue gas (112°C) is extracted and exported to a heating
316 network. The flue gas contains 3528 kg/h of CO₂, of which 86% is due to the combustion of SCG
317 material.

318 **Validation of simulation results:** In this configuration, 5767 kg/h of SCG at 66% moisture
319 produces 283 kg/h of oil and 243 kg/h of biodiesel product. This corresponds to a yield of 0.86 kg
320 biodiesel per kg coffee oil, which is in close agreement with the average of yields found from
321 literature, i.e. 0.84 kg biodiesel/kg oil (Abdelaziz et al., 2020, 2019).

322 **3.5 Configuration II**

323 The process block diagram was illustrated in Figure 3.



324

325 Figure 3. Flow diagram of SCG biorefinery Configuration II.

326 **Pre-treatment:** To isolate lignin, Organosolv pre-treatment was selected as it is the only
 327 experimentally validated technique of SCG lignin isolation found in literature (Lee et al., 2019).
 328 The process conditions used are adopted from said study. The wet SCG feedstock (stream B1) does
 329 not undergo pre-drying as the high moisture content promotes solvent penetration into SCG during
 330 pre-treatment, therefore increasing delignification rates (Hochegger et al., 2019).

331 Solvents *n*-hexane and methanol are added to a reactor at 161°C and 1 bar, along with sulphuric
 332 acid as the catalyst. Under these conditions, lignin dissolves in methanol, while triglyceride and
 333 FFA are simultaneously extracted into *n*-hexane. The sulphuric acid catalyses the esterification of
 334 the extracted FFA, forming biodiesel. The solid residues consisting of mainly hemicellulose and

335 cellulose fibres are removed using a filter, forming stream B2. The remaining liquid mixture
336 (stream B3) is separated into stream B4 (containing solubilised lignin) and stream B5 (containing
337 mainly biodiesel and triglycerides) via centrifugation. Lignin and biodiesel recoveries of 56% and
338 62.4% are obtained from the Organosolv reactor (Lee et al., 2019). Oil extraction yield was taken as
339 78 wt% (Efthymiopoulos et al., 2018).

340 **Dilute acid pre-treatment and enzymatic hydrolysis:** The solid residues of stream B2 are slurried
341 in dilute sulphuric acid and treated at 163°C and 1 bar (Mussatto et al., 2011). Under these
342 conditions, hemicellulose is converted to its monomeric sugars. After pre-treatment, the solid
343 cellulose and hemicellulose hydrolysate are separated via centrifugation, after which the solids are
344 washed with water to remove any residual soluble sugars and fermentation inhibitors (sugar
345 degradation products). The water stream from this washing stage is merged with the hemicellulose
346 hydrolysate and the resulting stream is detoxified using a granular activated carbon column to
347 remove HMF and furfural formed (Nieder-Heitmann et al., 2019). The column output stream is sent
348 to a neutralisation reactor (38°C, 1 bar) where calcium hydroxide neutralises the remaining acid.
349 The resulting salt is precipitated and filtered out, leaving a hemicellulose sugar-rich stream (stream
350 B6) utilised in SA production. On the other hand, the solid cellulose residues (stream B7) from the
351 dilute acid pre-treatment are subjected to enzymatic hydrolysis at 48°C. Here, cellulase enzymes
352 convert the cellulose fibres into glucose monomers and small traces of soluble gluco-oligomers.
353 Solid residues of unreacted lignin, proteins, cellulose, and other compounds are removed from the
354 aqueous glucose stream (stream B8) via centrifugation.

355 **SA production from hemicellulose:** The detoxified, neutralised aqueous solution of hemicellulose
356 sugars (stream B6) is utilised in SA production using the bacteria *Actinobacillus succinogenes*
357 (Nieder-Heitmann et al., 2019). Like PHB production, growth and synthesis reactors are used. Both
358 reactors operate at 38°C. With 10% of stream B6 directed to the growth reactor, the remaining is
359 sent to the synthesis block, after passing through flash drums to concentrate the hydrolysate stream
360 to 200 g/L of sugars (Nieder-Heitmann et al., 2019).

361 The products of the synthesis reactor are filtered to remove cells and the liquid product stream sent
362 to an adsorption tower. An adsorption tower using resin NERCB 09 is used due to its high SA
363 selectivity and average recovery of 96% (Li et al., 2009). After adsorption, the adsorbent is easily
364 regenerated using NaOH solution. As SA reacts with the base to form sodium succinate, sulphuric
365 acid is added to the reactor, forming sodium sulphate and SA. A patented process involving
366 selective precipitation of sodium sulphate is applied to recover the SA (Fujita and Wada, 2008).
367 Finally, a distillation column removes water, producing the SA product stream B9.

368 **PHB production from cellulose:** A fraction of the glucose-rich stream B8 is diverted as feed for
369 the cellulase production plant at a rate of 3.9 kg/h of glucose per kg/h of cellulase required
370 (Humbird et al., 2011). The remaining glucose is used for PHB production. Although there exists a
371 variety of microbes with the ability to produce PHB intracellularly, the use of recombinant *E. coli*
372 was chosen as it produces PHB during both growth and synthesis phases, resulting in lower
373 residence times and capital costs (Nieder-Heitmann et al., 2019). The PHB production process
374 consists of growth and synthesis phases, product separation and purification. The glucose stream
375 from the enzymatic hydrolysis of cellulose is split into two streams, with 9.5% sent to the growth
376 reactor and the remaining to the synthesis reactor. Before these glucose streams enter the reactors,
377 they are either mixed with water or undergo evaporation to achieve glucose concentrations of 20
378 g/L and 700 g/L for the growth and synthesis reactors, respectively. The growth reactor produces a
379 population of microorganisms from a stock dormant culture, sugar, and ammonia for use in PHB
380 production in the synthesis reactor. The microbial cells are sent to the synthesis reactor, where
381 concentrated glucose (700 g/L) is fed for continued bacterial growth and intracellular PHB
382 production. A centrifuge separates the cells containing PHB from the fermentation broth. After a
383 water wash at 30°C, the cells are lysed in a blending tank at 30°C containing a 0.2 M NaOH
384 solution to release the PHB within the cells. The PHB crystallises upon exposure to the solution
385 (Nieder-Heitmann et al., 2019). Finally, the crystalline PHB is subjected to a final water wash and

386 centrifugation to remove any residual cell material before being atomized into particles using a
387 spray dryer, forming stream B10.

388 **Biodiesel production from oil:** Stream B4 is subjected to identical transesterification and biodiesel
389 purification processes as in Configuration I. However, *n*-hexane is first separated from the lipids
390 using a flash drum at 75°C and 0.06 bar. Biodiesel (stream B11) is produced.

391 **Production of aromatic compounds and acids from lignin:** The soluble lignin of stream B5 is
392 precipitated out via the addition of water and the solid lignin formed is separated using a filter. This
393 lignin and compressed air enter an oxidative depolymerisation reactor (160°C, 8 bar) (Fujita and
394 Wada, 2008; Li et al., 2009) where lignin reacts with oxygen to produce vanillin, vanillic acid,
395 formic acid, acetic acid, guaiacol, and acetovanillone (streams B12-B17), along with CO₂.
396 Unreacted lignin is removed via filtration and recycled back to the depolymerisation reactor to
397 maximise production of the high-value chemicals. The products in the liquid stream are isolated
398 using a novel hydrophobic membrane separator that has been shown to achieve perfect separation of
399 organic and aromatic compounds from aqueous mixture (IEA Bioenergy, 2021; Phillips et al.,
400 2019). The membrane unit thus was modelled using a separator block with perfect separation of
401 organic compounds. The products in the organic phase output stream of the membrane separator are
402 separated using a series of distillation columns under vacuum conditions to give product streams
403 B12-B17 of 98-99% purity.

404

405 **4 Results and Discussion**

406 The parameters and assumptions used in the scenarios were outlined in Section 4.1. Performance
407 analyses explored the influence of size, location and products on the economic and environmental
408 performance of a biorefinery, presented in Section 4.2. The biorefinery GHG emissions were
409 compared against that of conventional SCG management practices. Biorefinery performances were

410 also compared against that of conventional biodiesel. Section 4.3 provides insights on the future
 411 prospects of SCG biorefineries and recommendations for future studies.

412 **4.1 Parameters and Assumptions for Scenarios**

413 **Parameters for performance evaluation:** To determine economic performance, h was taken as
 414 8000 hours. For labour cost calculations (Equation (6)), Configuration I involved 3 processing steps
 415 and was considered automated, requiring one personnel per step. Configuration II had 6 processing
 416 steps, with 3 personnel required per step due to the complexity of the plant design. The EP and NPV
 417 were calculated using Equations (8)–(9), where the values of Np were 3 and 9 for Configuration I
 418 and II respectively. Price sensitivity analysis was performed (see Section 2) with price fluctuations
 419 ($\Delta\%$) of 10%. The unit product prices are summarised in Table D.1 of Appendix D. The cost of
 420 production of biodiesel (£/L) was calculated using Equation (14).

$$\text{Biodiesel cost} = \frac{\text{Total annual production cost}}{\text{Litres of biodiesel produced annually}} \times \frac{\text{Biodiesel revenue}}{\text{Total revenue}} \quad (14)$$

421 Net GHG values were calculated using Equation (10) and net heat and electricity consumptions
 422 were calculated by subtracting heat and electricity consumed from that produced by SCG
 423 combustion.

424 **4.2 Performance Analysis**

425 The parameters and key performance indicators of each scenario are summarised in Table 2, where
 426 negative values of net heat or electricity consumption represents net generation and subsequent
 427 export to heat networks or the electrical grid.

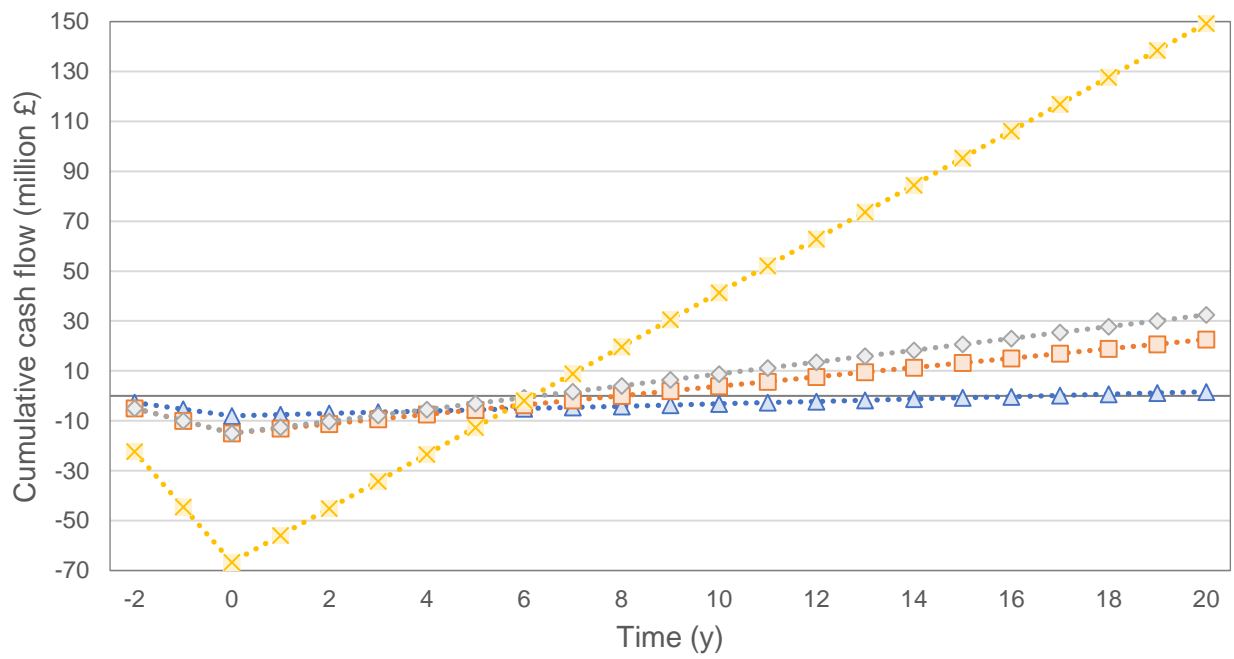
428 Table 2. Parameters and key performance indicators of the scenarios analysed.

	Scenario A	Scenario B	Scenario C	Scenario D
Brief Description	Config I London SCG Off-site	Config I Factory SCG Off-site	Config I Factory SCG On-site	Config II London SCG Off-site
Parameters				
Configuration	I	I	I	II

SCG feedstock source	London shops	Nestlé factory	Nestlé factory	London shops
SCG transport distance (km)	70	70	-	70
SCG feedstock temperature (°C)	20	20	80	20
SCG feedstock flowrate (t/y)	46136	105882	105882	46136
Technical Performance Indicators				
Biodiesel yield (kg biodiesel/t SCG processed)	42.10	42.10	42.10	33.60
Net heat consumption (MJ/t SCG processed)	- 3392.93	-3501.58	-3522.28	7575.38
Net electricity consumption (MJ/t SCG processed)	-185.12	-195.01	-195.01	144.84
Economic Performance Indicators				
Economic potential, <i>EP</i> (million £/y)	-0.46	0.12	0.62	2.97
Net present value, <i>NPV</i> (million £)	-3.90	-0.42	3.07	15.19
Payback period (y)	16.6	8.0	6.3	6.2
Cost of biodiesel production (£/L)	1.02	0.81	0.72	0.69
Environmental Performance Indicators				
GHG emissions (kg CO ₂ -eq./t SCG processed)	81.21	80.98	78.61	2233.00

429

430 Figure 4 presents the economic and environmental performance of the scenarios. The cumulative
431 cash flows of the scenarios are illustrated in Figure 4 (a), where *TCC* is evenly divided between
432 years -2 to 0. Positive and negative *NPV* values indicate profitable and loss-making projects,
433 respectively. The payback periods of Table 2 are the number of years required to recover the *TCC*
434 investment. These are represented by the x-axis intercepts on Figure 4 (a) and are a measure of the
435 plant's attractiveness as an investment. Revenues are categorised by product sales in Figure 4 (b)
436 and cost breakdowns of the scenarios are shown in Figures D.1–D.3. The results of the price
437 sensitivity analysis are illustrated in Figure 4 (c), with the legend indicating the classes of economic
438 risk. For example, '0 to -20%' economic risks referred to price combinations that resulted in a 0 to
439 20% decrease in *EP* relative to the *EP* generated from base prices. For Scenario A, none of the price
440 combinations resulted in a positive value for *EP*, while all the price combinations of Scenarios C
441 and D generated positive *EP*.

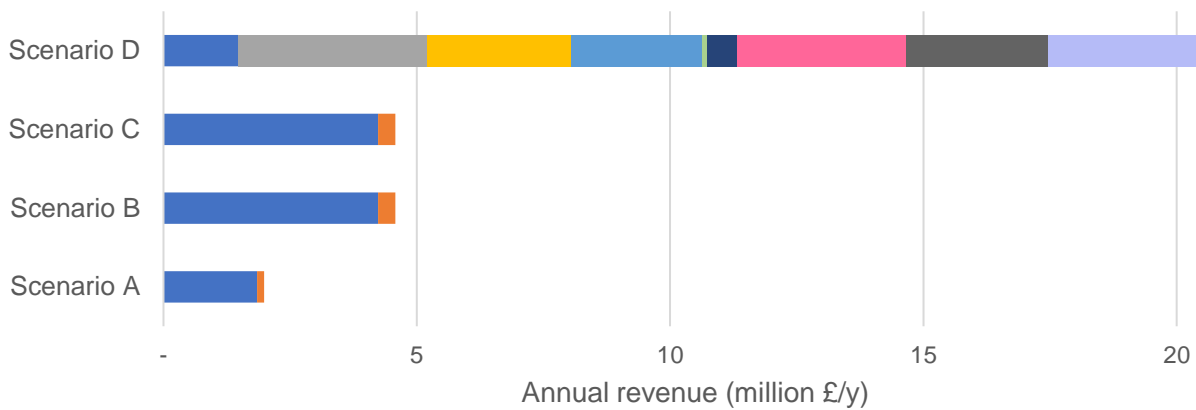


Scenario A Scenario B Scenario C Scenario D

442

443

(a)

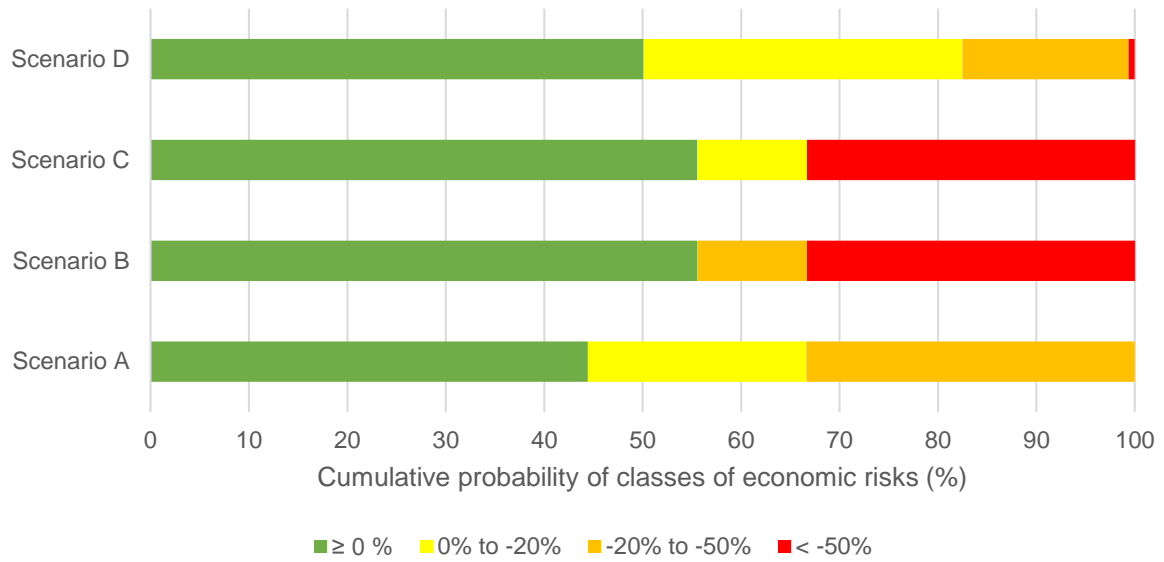


Biodiesel Electricity Vanillin Vanillic acid Guaiacol
Acetic acid Formic acid Acetovanillone Succinic acid PHB

444

445

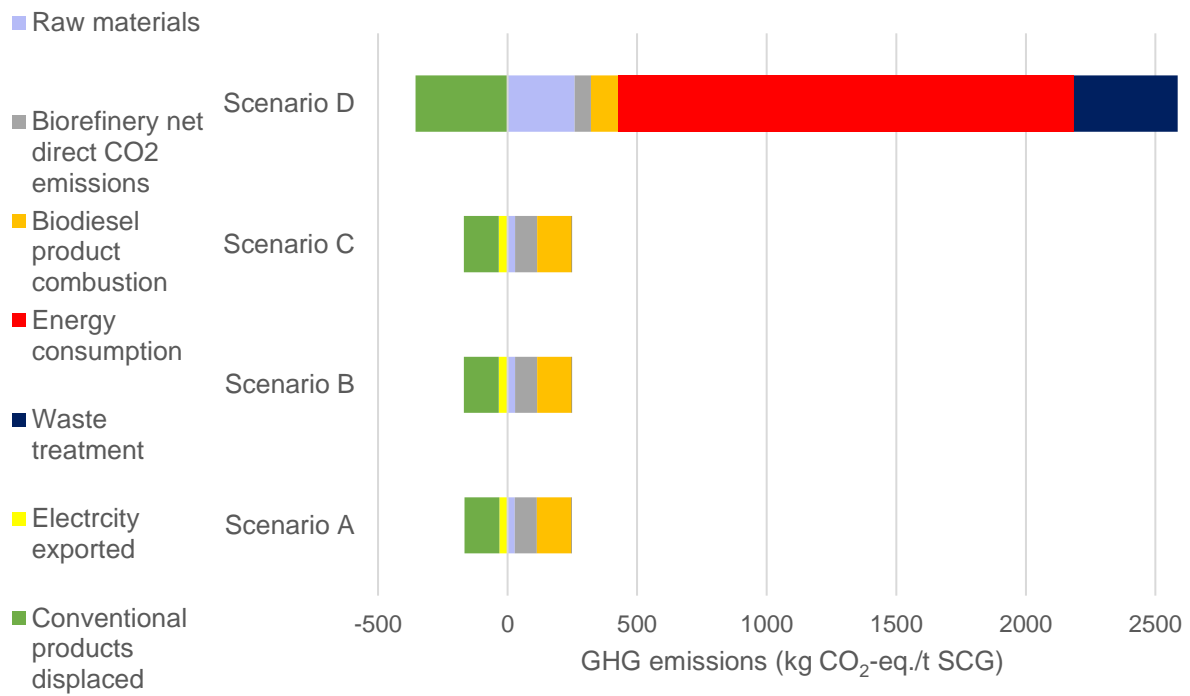
(b)



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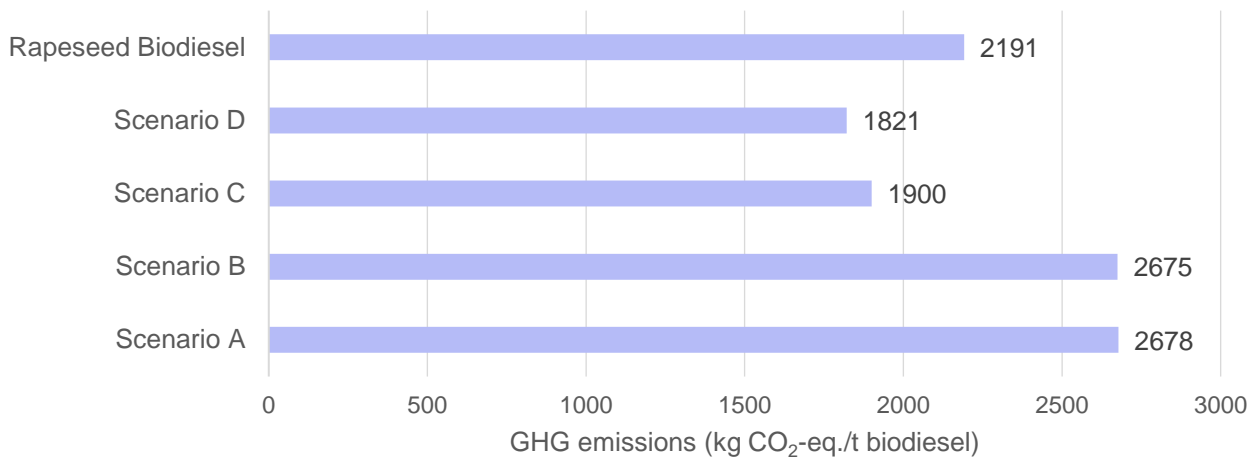
(c)



448

449

(d)



450

451

(e)

452 Figure 4. Economic and environmental performance of scenarios analysed. (a) Cumulative cash
 453 flows for the plant's 20 years of operation; (b) Classification of revenues by product sales; (c)
 454 Likelihood of different classes of economic risk; (d) Classification of GHG emissions per tonne of
 455 SCG processed; and (e) GHG emissions per tonne of biodiesel produced.

456

457 **Plant size:** The impact of increasing biorefinery size was investigated by comparing the
 458 performances of Scenario A and B, with SCG feedstock flowrates of 46136 and 105882 t/y,
 459 respectively. Scenario A generates a negative *NPV* value of -£3.9 million, implying that this
 460 scenario is not economically compelling. Increasing the plant size by 2.3 times to that of Scenario B
 461 still resulted in an economically unviable plant, with a *NPV* of -£0.42 million (Table 2). However,
 462 the increase in *NPV* along with the reduction in payback period from 16.6 to 8.0 years (Figure 4 (a))
 463 indicates that Scenario B is a more attractive investment than Scenario A. The cost of biodiesel
 464 production was reduced by 18% from 0.88 to 0.72 £/L (Table 2). This improved economic
 465 performance aligns with previous hypotheses that larger scale of SCG biorefineries is required to
 466 result in an economically viable production (Giller et al., 2017). Besides that, a price sensitivity
 467 analysis of both scenarios (Figure 4 (c)) reveals that Scenario B has a 11% greater likelihood of
 468 $\geq 0\%$ economic risks when product prices fluctuate from their base levels. A $\geq 0\%$ economic risk
 469 refers to an increase or no change in *EP* relative to the *EP* generated at base prices, thus requiring
 470 no modification to the biorefinery processes. The likelihood of $< -50\%$ economic risks rose from

471 0% (Scenario A) to 33% (Scenario B). This type of economic risk would greatly reduce *NPV* and
472 thus would require plant process modifications to minimise its large negative economic impact.
473 Generally, the greater the likelihood of $< -50\%$ economic risks, the higher the economic risk of the
474 scenario. Thus, increasing the biorefinery size results in an overall rise in the plant's economic risk.

475 On the other hand, environmental performance (see Figure 4 (d)) does not improve by increasing
476 biorefinery size, with the GHG emissions of Scenario B a mere 0.3% lower than that of Scenario A.
477 Biodiesel yield per tonne of SCG processed is not affected while the net heat and electricity
478 generated per tonne of SCG increase with biorefinery size.

479 **Biorefinery location:** Scenarios B and C were compared to examine the impact of biorefinery
480 location relative to the SCG source. Locating the biorefinery within Nestlé's Derbyshire coffee
481 factory site results in significant improvement in economic performance, with *NPV* growing by 9.3
482 times to £3.1 million, a 1.7-year reduction in payback period and a 11.1% reduction in biodiesel
483 production cost. These findings demonstrate the significance of a centralised processing facility on
484 the economic performance of SCG valorisation, as concluded by Kookos (2018). Economic risk
485 also decreases, with around a tenth of price combinations generating economic risks of 0% to
486 -20% instead of -20% to -50% . However, there remains a 33% likelihood that 10% product price
487 fluctuation results in $< -50\%$ economic risk, indicating that the economic viability of Scenario C is
488 still vulnerable to market price fluctuations. Besides that, the omission of SCG transport results in a
489 30% reduction in raw material cost and a 3% reduction in GHG emissions. While biodiesel yield
490 and net electricity consumption remain unchanged, net heat generated increases slightly by 0.5% as
491 the SCG feedstock enter at 80°C , resulting in lower SCG drying heat requirements.

492 **Biorefinery products:** The impact of producing high-value chemicals was assessed by comparing
493 Scenarios A and D. Scenario D has excellent economic performance, with a *NPV* of £15.2 million
494 and a payback period of 6.2 years. The cost of biodiesel production decreases by 4% from 0.72 £/L
495 to 0.69 £/L due to biodiesel comprising a much smaller proportion of total revenue and use of

496 Equation (14). This result supports the IEA's proposition that producing high-value products
497 alongside biofuels would lower biofuel production costs (van Ree et al., 2012). As illustrated in
498 Figure 4 (b), almost 90% of the annual revenue generated by Scenario D is evenly distributed
499 between sales from vanillin, vanillic acid, guaiacol, acetovanillone, SA and PHB. This even
500 distribution of revenue across many products leads to the reduced economic risk seen in the results
501 of the sensitivity analysis in Figure 4 (c). In contrast, Scenario A has much higher economic risk
502 due to biodiesel sales constituting 93% of its revenue. Thoppil and Zein (2021) also found the *NPV*
503 of SCG biorefinery to be highly sensitive to product prices when products are of limited range and
504 low value.

505 However, the economic performance of Scenario D is at the cost of greater environmental impact,
506 with GHG emissions increasing by 27.5 times to 2233 kg CO₂-eq./t SCG (Figure 4 (d)). Steam
507 generation and electricity usage constitute 52% and 18% of the total emissions from the plant,
508 respectively. These emissions have been grouped and labelled as 'energy consumption' in Figure 4
509 (d). For its GHG emissions/t SCG to match that of Scenario A, the energy consumption of Scenario
510 D would have to be reduced by at least 22%. As Scenario D does not involve on-site energy
511 recovery via combustion, it results in net electricity and heat consumption 3.2 and 1.8 times that of
512 Scenario A. Besides that, Scenario D results in a biodiesel yield around 20% less than Scenario A
513 due to the lower biodiesel yields of the Organosolv pre-treatment.

514 **Conventional SCG management practices:** The environmental impacts of Scenarios A and D
515 were compared against that of the two most widespread SCG management practices in the UK:
516 EfW and landfilling. Schmidt et al. (2020) found the GHG emissions of SCG EfW and landfill
517 facilities with energy recovery to be -435 kg CO₂-eq./t SCG and 525 kg CO₂-eq./t SCG,
518 respectively. Neither Scenario A nor D have GHG emissions as low as EfW practices. Scenario A
519 produces around 85% less GHG emissions than landfilling while Scenario D produces more than 4
520 times the GHG emissions of landfilling. Note that Scenarios B and C were excluded from this
521 comparison due to the similarity of their GHG emissions with that of Scenario A.

522 **Conventional biodiesel production:** The GHG emissions of Scenarios A to D were compared
523 against that of conventional biodiesel production in Europe. Rapeseed oil is the dominant feedstock
524 for EU biodiesel production, constituting 39% of total production in 2018 (Phillips et al., 2019).
525 The GHG emissions generated from rapeseed biodiesel produced via esterification was reported by
526 Pehnelt and Vietze (2012), whose study excluded emissions from the transportation of the biodiesel
527 product to the consumer, biodiesel combustion and waste treatment. A functional unit of 1 tonne of
528 biodiesel production was used. Adopting identical system boundaries and functional unit, the GHG
529 emissions generated by biodiesel production was calculated by summing the emissions from SCG
530 collection, Organosolv pre-treatment and transesterification. For the collection and pre-treatment,
531 the GHG emissions generated from biodiesel production were calculated using Equation (15),
532 where ‘Process GHG’ refers to the total GHG emissions of each process.

$$\text{GHG from biodiesel production} = \text{Process GHG} \times \frac{\text{Biodiesel revenue}}{\text{Total revenue}} \quad (15)$$

533
534 The GHG emissions of the scenarios were compared against that of rapeseed biodiesel as shown in
535 Figure 4 (e). The GHG emissions in Scenario C (1900 kg CO₂-eq./t biodiesel) are 29% lower than
536 that of Scenario B (2678 kg CO₂-eq./t biodiesel) as it excludes transportation of the 23.8 tonnes of
537 SCG required to produce 1 tonne of biodiesel. Besides that, Scenarios C and D produce GHG
538 emissions that are 13% and 17% lower than rapeseed biodiesel.

539 The relatively low emissions of Scenario D are due to the exclusion of waste treatment emissions
540 (which constitute 18% of total GHG emissions/t SCG) and use of Equation (15) (as biodiesel
541 revenue represents just 7% of total revenue). Although the GHG emissions/t biodiesel appear
542 promisingly low, Figure 4 (d) shows that total GHG emissions generated by Scenario D are 7 times
543 greater than the emissions credited from displacing the use of conventional products. Thus,
544 producing these high-value chemicals (e.g., vanillin, PHB) via the SCG biorefinery results in
545 greater GHG emissions than conventional production methods.

546 According to the IEA, the production cost of conventional biodiesel ranges from 0.66–0.86 £/L
547 (International Energy Agency, 2012). All the scenarios apart from Scenario A result in biodiesel
548 production costs that are within this range. The results from this comparison indicate that SCG
549 biorefineries could potentially be used to meet growing biodiesel demands.

550 **4.3 Insights and Recommendations**

551 From the economic and environmental evaluations performed in Section 4.2, Scenario C was found
552 to be the most promising SCG biorefinery strategy. Although its *NPV* of £3.1 million is around 5
553 times lower than that of Scenario D, its GHG emissions (79 kg CO₂-eq./t SCG processed or 1900 kg
554 CO₂-eq./t biodiesel) are 28 times lower than Scenario D, 7 times lower than SCG landfilling (525
555 kg CO₂-eq./t SCG) and 13% lower than that of conventional rapeseed biodiesel (2191 kg CO₂-eq./t
556 biodiesel). It also has a biodiesel cost of production of 0.72 £/L, which is within the range estimated
557 by IEA (0.66–0.86 £/L). Furthermore, a practical advantage of using SCG from a coffee factory is
558 a more consistent feedstock flowrate and composition than SCG from coffee establishments,
559 resulting in fewer changes in operating conditions and product output. However, the economic
560 viability of Scenario C is sensitive to product price fluctuations, with 55.6% and 33.3% likelihoods
561 of $\geq 0\%$ and $< -50\%$ economic risks, respectively (Figure 4 (c)). Nonetheless, a positive *EP* is
562 maintained for price fluctuations within 10% of the base price.

563 Theoretically, the supply of the net heat generated to heat networks would represent an opportunity
564 to further reduce the biorefinery's carbon emissions through application of carbon credits. However,
565 the Derbyshire coffee factory considered in this study is one of more than 20 Nestlé coffee factories
566 worldwide currently incinerating the SCG it produces to meet the factory's heat energy
567 requirements ("Grounds for sustainability: coffee for energy, fuel and a cleaner world," 2013).
568 Future studies on the valorisation of SCG from instant coffee factories must thus account for the
569 economic and environmental impact of diverting SCG to a biorefinery and away from combustion
570 to generate the heat requirements of the factory. Including this in the analysis would likely have
571 negative implications on both the economics and emissions of the SCG biorefinery. However, the

572 biorefinery strategy could still represent a more profitable method of utilising SCG waste than
573 combustion. As size was found to be an important factor affecting the economic performance and
574 risk of a SCG biorefinery, different coffee factory sizes should be explored in these studies.

575 Additionally, the environmental evaluations of this study only considered GHG emissions. The
576 environmental performances of the scenarios may differ upon assessment of other categories such
577 as acidification potential and human toxicity potential. Other environmental impact categories
578 should also be evaluated in future studies for a more holistic view of SCG biorefinery
579 environmental performance.

580 Oil extraction and PHB production were identified as the processes with the largest costs in
581 Scenarios A and D (see Figure D.3 of Appendix D). *n*-hexane and cellulase represent 25% and 17%
582 of the total annual costs of Scenarios A and D, respectively. Thus, further studies on biodiesel and
583 PHB production from SCG should aim to reduce usage of *n*-hexane and cellulase to maximise
584 potential cost reductions. The technologies used in Scenario C must also be scaled to pilot plant
585 levels to verify technical feasibility of the SCG biorefinery strategy. The accuracy of this study's
586 results depends heavily on the financial investments and engineering capability required to scale
587 these processes up economically.

588

589 **5 Conclusions**

590 This study investigates the future prospects of SCG biorefineries by assessing four SCG biorefinery
591 scenarios in terms of their economic and environmental performances. The parameters altered
592 between scenarios include biorefinery size, design configuration and location relative to the SCG
593 source. SCG was either obtained from London coffee establishments or a coffee factory in
594 Derbyshire. Two biorefinery configurations were used, with one producing biodiesel and electricity
595 (Configuration I) and the other biodiesel with a range of high-value chemicals (Configuration II).

596 Overall, the results of this study indicate that future SCG biorefineries are likely to be of large scale
597 and located on the site of an instant coffee factory. Heightened growth in demand for advanced
598 biofuels coupled with pressures to reduce factory emissions mean that the waste-to-fuel biorefinery
599 pathway could be regarded as an economically and environmentally attractive additional value
600 chain for coffee factories. Provided that the relevant technologies can be successfully scaled up,
601 production of multiple high-value chemicals is likely to be incorporated into SCG biorefineries due
602 to attractive economic returns. Implementation of sustainable on-site energy generation will be
603 crucial to minimise the high GHG emissions of the plant. Finally, SCG biorefineries have the
604 potential to meet growing biodiesel demand, with similar production costs and lower GHG
605 emissions when compared to conventional biodiesel.

606

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611 **References**

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