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Life cycle environmental sustainability of valorisation routes for spent coffee grounds: From waste to resources

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ABSTRACT

Spent coffee grounds (SCGs) have a potential to be used as a feedstock for higher value-added products, such as biodiesel. However, the environmental implications of the valorisation of SCGs are largely unknown. This study evaluates the life cycle environmental impacts of utilising SCGs for biodiesel production in comparison with the widely used disposal of SCGs as a waste stream: incineration, landfilling, anaerobic digestion, composting and direct application to land. The scope is from cradle to grave and the functional unit is defined as 'treatment of 1 tonne of SCGs'. The results show that the most environmentally sustainable option is incineration of SCGs, with net-negative impacts (savings) in 14 out of 16 categories, followed by direct application of SCGs to land with 11 net-negative impacts. Biodiesel production is the least sustainable option with the highest impacts in 11 categories, followed by composting. The paper also demonstrates that following various waste hierarchy and resource valorisation guidelines instead of a life cycle approach could lead to a choice of environmentally inferior SCG utilisation options. Therefore, these guidelines should be revised to ensure that they are consistent and underpinned by life cycle thinking, thus aiding sustainable resource management in a circular economy context.

1. Introduction

The development of global initiatives promoting value-added creation of waste streams, also known as waste valorisation, has become one of the main strategies for dealing with food waste while increasing resource efficiency and reducing environmental pressures (EC, 2017a; WRAP, 2018a). The United Nations (UN), the World Economic Forum (WEF) and the European Union (EU) advocate a transition to a more circular economic model. In this model, resources, materials and products are kept within systems for longer to increase their value while reducing waste and environmental impacts, creating jobs and promoting sustainable growth (UNEP, 2017; EC, 2017b; WEF, 2017). Within this context, the EU has developed the Bio-economy Strategy, which promotes the use of innovative bio-technological solutions for converting currently-discarded renewable resources, such as food waste, into value-added products, including bio-energy, food and animal feed (EC, 2017b).

Coffee, the second largest beverage consumed worldwide after tea (Scully et al., 2016), is an example of a product with a high rate of unavoidable waste at the point of consumption, generating 1.88 kg of spent coffee ground (SCGs) per kg of coffee beans used (Cameron and O'Malley, 2016). SCGs are the primary unavoidable (inedible) waste from ground roasted coffee (Esquivel and Jiménez, 2012), which is produced mainly from two sources: the soluble (instant) coffee industry and consumption in catering outlets (e.g. cafes and restaurants) and homes (Scully et al., 2016).

The UK is the fifth largest market for coffee in Europe, with annual imports of 300 kt of green beans, roasted and instant coffee (ICO, 2018). Excluding the instant coffee waste (industrial waste produced in the country of origin), the UK generates an estimated 256.8 kt of SCGs a year (see Table S1 in the Supplementary Information (SI)). Currently, SCGs are considered a waste and are treated as such with the majority being landfilled or incinerated (Quested and Parry, 2017). There are no specific guidelines to handle and manage SCGs and are

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considered as food waste (WRAP 2018b).

However, SCGs show a potential for valorisation in the bio-economy context. They are an abundant and a low-cost resource that can be utilised through simple valorisation routes, such as waste-to-energy (e.g. incineration with energy recovery, biomass logs and briquettes) as well as for more complex high-end-value products, such as enzymes and aromas used in the food, cosmetic and pharmaceutical industries (Karmee, 2017). However, despite the extensive research exploring potential valorisation options for SCGs, little is known about the prospective environmental benefits that these routes could offer. This is in contrast to the abundance of studies on environmental impacts of coffee production (e.g. Salinas, 2008; Salomone, 2003) and its consumption (e.g. Hassard et al., 2014; Hicks, 2018; Humbert et al., 2009).

In terms of waste-to-energy valorisation routes, Itten et al. (2011) assessed the conversion of SCGs into briquettes for heating and compared them with the equivalent products from other biomass sources, including horse dung, poultry litter, pig slurry and olive pomace. The authors concluded that all these biomass sources had a lower global warming potential than fossil fuels but higher than wood. Furthermore, the authors explored the trade-offs between greenhouse gas (GHG) emissions and other health-related impacts, showing that the biomass sources had much higher emissions of heavy metals, particle and NO_x than the fossil and wood fuels.

A couple of studies considered utilisation of SCGs for production of biodiesel. One of these (Kookos, 2018) carried out a gate-to-gate techno-economic analysis and estimated the carbon footprint of producing SCGs biodiesel in the conventional two-step transesterification process. The results suggested that biodiesel had net-negative GHG emissions (savings) due to the biogenic carbon sequestered by the SCGs. Also focusing on the production stage only, the second study (Tuntiwiwattanapun et al., 2017) considered the same process but in comparison with a new one-step esterification method. The authors found that the conventional process had lower energy consumption and the climate change impact than the one-step alternative, but higher toxicity-related impacts and land use.

As far as the authors are aware, there are no comprehensive studies that analysed and compared the life cycle environmental impacts of different SCGs management practices and valorisation routes. Therefore, this work evaluates the implications of using SCGs for biodiesel as one of the high value-added products. This valorisation route is compared to the following management methods currently used to deal with the SCG waste: incineration, landfilling, anaerobic digestion, composting and direct application to land. The paper also aims to find out if the most sustainable options identified through the life cycle perspective correspond to those recommended in various waste valorisation hierarchies in an attempt to improve the consistency across different methods.

2. Methods

The life cycle assessment (LCA) study has been conducted according to the ISO 14040/44 guidelines (ISO, 2006a; 2006b), following an attributional approach. The assumptions and data are detailed in the following sections, starting with the definition of the goal and scope in Section 2.1. The inventory data and assumptions for each of the six SCG management routes are detailed in Section 2.2. An overview of the impact assessment method applied in the study is provided in Section 2.3.

2.1. Goal and scope

The main goal of this study is to estimate and compare the environmental impacts associated with different SCGs management and valorisation routes as follows:

- biodiesel production;
- anaerobic digestion with electricity production and digestate use in agriculture;
- composting;
- direct application of SCGs to land;
- incineration with electricity and heat generation; and
- landfilling with biogas recovery for electricity generation.

Apart from biodiesel, all of the above routes are used mainly to manage waste rather than recover valuable components (e.g. oils, enzymes or aromas) as precursor for high value-added products (e.g. cosmetics or food supplements). However, given that they all recover useful products (e.g. heat or electricity), and recognising their potential to be considered in the future as valorisation routes, they are referred to as such in the rest of the paper.

These valorisation routes have been selected based on two aspects: a) current waste management of SCGs; and b) available information. At present, SCGs are treated together with food waste; however, there are already initiatives, mainly private, whereby SCGs are collected separately and transported to facilities to produce liquid and solid biofuels (Bio-bean Limited, 2019). Similarly, SCGs have been used as soil enhancer via compost and direct application (Starbucks, 2015). Furthermore, biodiesel has been one of the most successful initiatives for valorising SCGs so far (Bio-bean Limited, 2019; BBC, 2017).

The functional unit is defined as ‘treatment of 1 tonne of SCGs’. This is congruent with the fact that SCGs currently represent waste and are managed as such. As shown in Fig. 1, the scope of the study is from cradle to grave, comprising the following stages:

- transport: lorry transport of SCGs from the source to the treatment facility, considering a generic distance of 45 km;
- construction: infrastructure for the treatment plants;
- operation: energy, other utilities and materials used for SCG treatment, emissions associated with the treatment and waste management of residues from the process; and
- use of products produced in each treatment option.

The SCGs are assumed to be collected from catering outlets and industrial sites, separately from food waste streams. This assumption is based on above-mentioned initiatives by some companies (Bio-bean Limited, 2019).

As also indicated in Fig. 1, based on the ISO 14040/44 guidelines, the systems have been credited for their co-products as follows:

- biodiesel: for glycerine and fossil diesel production and combustion;
- anaerobic digestion: for electricity and fertilisers;
- composting and direct application of SCGs to land: for fertilisers; and
- incineration and landfilling: for electricity and/or heat.

The impacts of coffee production and consumption are excluded, and, therefore, SCGs are considered impact-free, in accordance with common LCA practice (Ekvall et al., 2007; EC, 2009).

2.2. Inventory data

The inventory data are discussed below, with further details provided in Table S2 in the SI. Ecoinvent 3.3 database has been used for the background data (Moreno Ruiz et al., 2016) assuming UK conditions.

2.2.1. Biodiesel production

Biodiesel is produced from SCGs using a two-step transesterification (TE) process with the first step involving oil extraction (using solvents) and the second converting oil into biodiesel via TE. Despite its hazardous characteristics, n-hexane is the most common solvent used for the first step - oil extraction (Tuntiwiwattanapun et al., 2017), while methanol is commonly used for the second step - TE. To reduce the

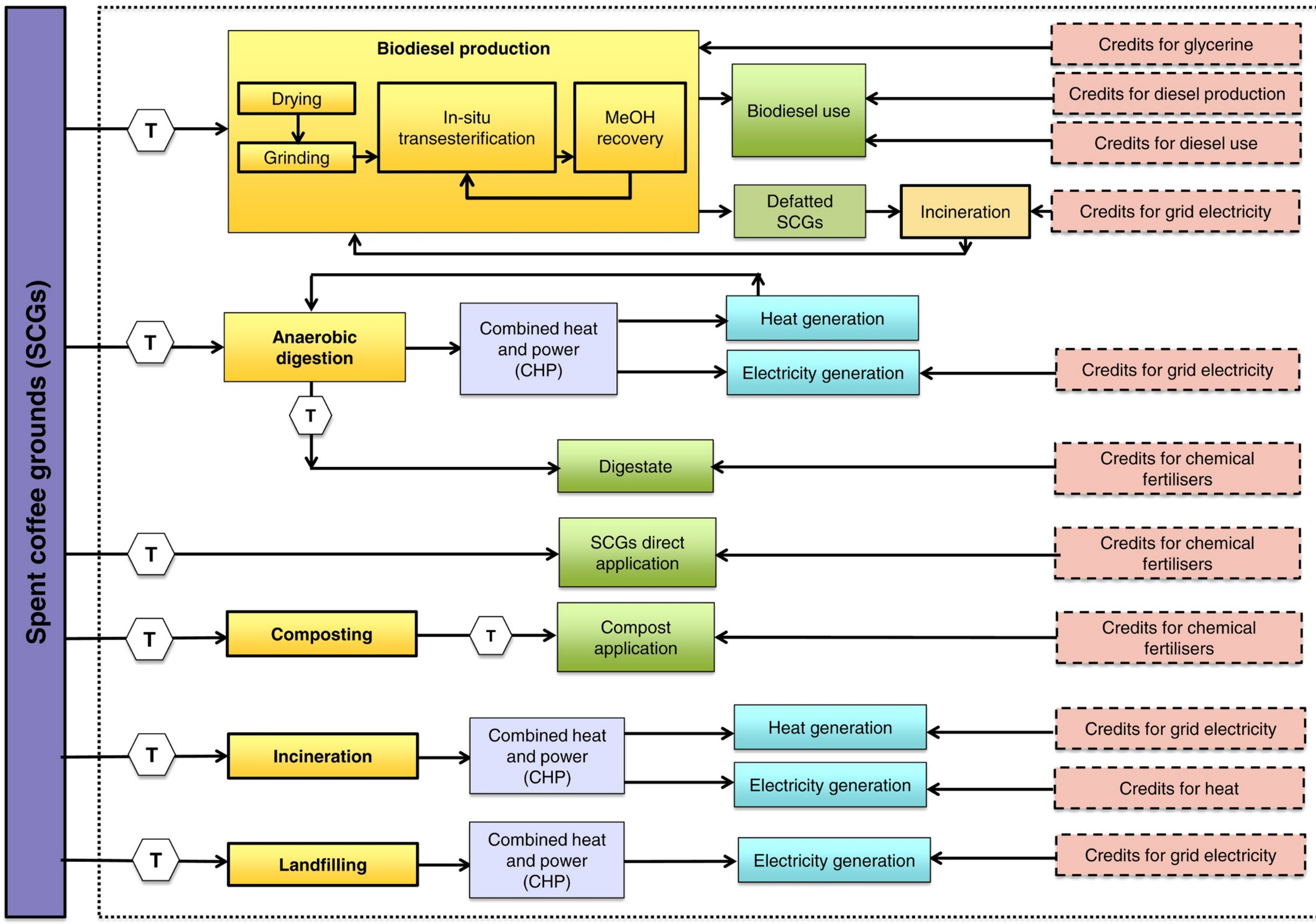


Fig. 1. System boundaries considered in the study (SCGs: spent coffee grounds; MeOH: methanol; T: transport).

hazards and economic costs, a new, one-step process, known as in-situ TE, has been recently proposed. This process uses methanol for both extraction and TE, hence avoiding the use of solvents like n-hexane. In-situ TE has a biodiesel yield of up to 96%. It also reduces the complexity and scale of the biodiesel production process, making it more attractive for smaller-scale applications (Tuntiwiwattanapun et al., 2017; Najdanovic-Visak et al., 2017). For these reasons, this study considers this new in-situ TE process. As there are no commercial plants currently in operation, the inventory data are based on the conceptual engineering design scaled up to an industrial level (Piccinno et al., 2016).

As can be seen in Fig. 1, this process involves first drying and grinding of SCGs, followed by in-situ TE to produce biodiesel and glycerine. The defatted SGCs remaining after TE are incinerated in a combined heat and power (CHP) plant to generate the heat and electricity needed in the process, including for the recovery of methanol. The energy efficiency of common CHP plants using biomass feedstock (45% for heat and 15% for electricity) has been assumed; the heating values of defatted SCGs can be found in Table S3 in the SI. The inventory data are summarised in Table 1.

The energy required for drying and grinding has been determined based on Piccinno et al. (2016) and Tuntiwiwattanapun et al. (2017). The excess heat and electricity not used in the process (see Table 1), is exported to the grid, crediting the system for the avoided impacts of high-voltage electricity and heat from natural gas. The system has also been credited for displacing fossil-derived glycerine (93% purity) (Kaewcharoensombat et al., 2011). Finally, the system has also been credited for the avoided impacts of fossil diesel production and use, based on their respective energy content (39.6 MJ/kg biodiesel and 45.5 MJ/ kg fossil diesel (Engineering ToolBox, 2009).

2.2.2. Anaerobic digestion (AD)

The AD system is based on facilities treating generic food waste (Slorach et al., 2019) as there are no AD plants for SGCs alone. However, the biogas production has been calculated considering the specific composition of the coffee waste. Data for the biogas production are based on Girotto et al. (2017) who reported an average yield of 360 m^3/t of volatile solids. A 2% leakage of biogas has been assumed (Slorach et al., 2019). As indicated in Table 2, the anaerobic digester considered here has a capacity of 2500 m^3, able to treat up to 25,000 t of SCGs under mesophilic conditions and to produce 196 kWh of electricity per tonne of waste in a CHP plant (Slorach et al., 2019). The co-produced heat is used in the system while the electricity is exported and credited to the system. The data for construction of the AD facility and CHP plant have been sourced from Ecoinvent 3.3 (Moreno Ruiz et al., 2016).

The digestate is transported to fields (25 km) where it is used as fertiliser (Saer et al., 2013). The nitrogen-based emissions have been modelled based on Nicholson et al. (2016), considering that 40% of the nitrogen in the digestate is emitted to the air as ammonia and 0.45% as nitrous oxide, while 15% leaches as nitrates (NO_2). The credits for the displacement of chemical fertilisers are based on recommendations in Slorach et al. (2019), assuming the displacement of 40% of ammonium nitrate as nitrogen-based fertiliser and 100% displacement of both phosphorous- and potassium-based fertilisers (phosphorus oxide and potassium oxide, respectively). Due to a lack of data on the digestate composition from SCGs, an average N-P-K composition from food waste

Table 1
Inventory data for biodiesel production from spent coffee grounds (SCGs) via in-situ transesterification.

Inputs and outputs	Unit (per t SCGs)	Amount	Source
Drying			Own calculations based on Piccinno et al. (2016)
Inputs			
Heat	MJ	600	
SCGs (wet matter, 49.3% moisture)	kg	1000	
Outputs			
SCGs (dry matter, 1% moisture)	kg	507	
Waste water	kg	493	
Grinding			Tuntiwiwattanapun et al. (2017)
Inputs			
Electricity	MJ	0.16	
SCGs (dry matter)	kg	507	
Outputs			
SCGs (dry matter)	kg	507	
Biodiesel production			Modelling based on Najdanovic-Visak et al. (2017) following Piccinno et al. (2016).
Inputs			
SCGs (dry matter)	kg	507	
Sodium hydroxide (catalyst)	kg	11.1	
Methanol	kg	632.5	42.5% reduction in methanol use from recovery of un-reacted methanol, based on Kaewcharoensombat et al. (2011)
Heat	MJ	25	
Steam	MJ	624.4	Methanol recovery uses 8.3 MJ/kg biodiesel and purification process 1.14 MJ/kg biodiesel (Varanda et al., 2011; Tuntiwiwattanapun et al., 2017).
Water	kg	206.856	3 kg/kg oil (Kaewcharoensombat et al., 2011)
Outputs			
Biodiesel	kg	66.15	
Defatted SCGs	kg	438	
Glycerine	kg	7.5	93% purity (Kaewcharoensombat et al., 2011)
Incineration of defatted SCGs			
Inputs			
Defatted SCG	kg	438	
Outputs			
Electricity (credits)	MJ	599	Own calculations; efficiency 15% (Ecoinvent 3.3)
Heat (credits)	MJ	1176	Own calculations; efficiency 45% (Ecoinvent 3.3)

Table 2
Inventory data for anaerobic digestion of spent coffee grounds (SCGs).

Parameter	Unit (per t SCGs)	Amount	Source
Facility capacity	t/yr	25,000	Scholes and Areikin (2014)
Electricity consumption	kWh	23	Slorach et al. (2019); Bernstad and la Cour Jansen (2011)
Heat consumption	kWh	82	Slorach et al. (2019)
Digestate production	t	0.82	Ecoinvent 3.3 (Moreno Ruiz et al., 2016)
Biogas production	m^3	133.2	Girotto et al. (2017)
Biogas leakage	%	2	Slorach et al. (2019)
Electricity production	kWh	196	Slorach et al. (2019)

has been assumed based on Bernstad and la Cour Jansen (2011). Table S4 in the SI details the composition of the digestate used as fertiliser and the emissions associated with it.

2.2.3. Industrial composting

An open-air composting facilities with turn windrows is considered here as one of the most common plants in the UK and elsewhere (Compost Certification Scheme, 2019). Composting is assumed to be carried out with the rest of food waste as there are no large-scale dedicated facilities for SCGs. At the plant, the waste is decomposed using multi-tunnel technology (Martínez-Blanco et al., 2019). In the decomposition process, SCGs remain in the tunnels, with forced aeration and irrigation used to aid the process. The decomposed SCGs are piled up and periodically turned to promote aeration. Finally, similar to the digestate from anaerobic digestion, the matured compost is transported to fields (25 km) to be used as fertiliser (Saer et al., 2013).

The composting system has been modelled based on data in Martínez-Blanco et al. (2009) and adapted to UK conditions using Ecoinvent 3.3 (Moreno Ruiz et al., 2016). Data for nutrient composition of the compost have been sourced from Gomes et al. (2013) and adapted to the specific characteristics of the SCGs considered in this study (see Table S4 in the SI). The emissions from applying composted SCGs are based on Nicholson et al. (2016) and the displacement of phosphorous- and potassium-based chemical fertilisers on Slorach et al. (2019). Nitrogen-based fertiliser has been modelled according to Bernstad and la Cour Jansen (2011), considering the displacement of 30% of ammonium nitrate. Table 3 shows the inventory data for the composting treatment while the details of N-P-K fertiliser displacement and the emissions from the use of composted SCGs can be found in Table S4 in the SI.

Table 3
Inventory data for industrial composting of spent coffee grounds (SCGs)[a]. Sourced from Ecoinvent 3.3 (Moreno Ruiz et al., 2016).

Parameters	Unit (per t SCGs)	Amount
Inputs		
Electricity[b]	MWh	0.03
Diesel[b]	m^3	4.45×10^{-3}
Water[b]	m^3	0.27
Outputs		
Compost[b]	T	0.14
NH_3[b]	kg	0.11
CH_4 (biogenic)[b]	kg	0.38
VOCs[b]	kg	1.21
N_2O[b]	kg	0.02
H_2S[c]	kg	0.08

[a] Emissions related to the use of compost are given in Table S4 in the SI.
[b] Volatile organic compounds. Sourced from Martínez-Blanco et al. (2009).
[c] Sourced from Ecoinvent 3.3 (Moreno Ruiz et al., 2016).

Table 4
Inventory data for incineration and landfilling of spent coffee grounds (SCGs)[a].

Parameter	Unit (per t SCGs)	Incineration[b]	Landfilling
Consumables			
Ammonia	g	549.6	10.3
Sodium hydroxide	g	365.2	0.5
$FeCl_3$	g	–	89.4
$FeSO_4$	g	–	65.3
Other chemicals[c]	g	24.1	0.94
Auxiliary fuel (natural gas)	MJ	53.6	1.8
Auxiliary fuel (light fuel oil)	MJ	–	0.6
Biogas			
Utilised	MJ	–	1333
Vented	MJ	–	769
Flared	MJ	–	281.9
Net energy generated (exported to the grid/heating)			
Electricity	MJ	2644.6	471
Heat	MJ	137.2	–
Waste			
Waste heat	MJ	10,486.2	
Landfill leachate	l	–	2500
Air emissions			
NOx	g	346.6	66.9
CO	g	222.9	20.9
N_2O	g	46	18.1
Dust, particulates	g	20.2	5.9
Cyanide	g	9.8	0.2
NH_3	g	8.6	6.6
CH_4 (biogenic)	g	6.4	17,016.8
SO_2	g	3.9	32.2
Phosphorous	g	0.9	0.009
Heavy metals	g	0.003	0.002
Other inorganic emissions	g	1	0.3
NMVOC[d]	g	–	0.4

[a] Modelled using the Ecoinvent tool for MSW sanitary landfills and incineration plants (Ecoinvent, 2008) based on the specific characteristics of SCGs (Table S3 in the SI).
[b] A weighted average, based on the 80%:20% share of electricity-only and CHP incineration plants.
[c] Modelled as generic inorganic chemicals sourced from Ecoinvent.
[d] Non-methane volatile organic compounds.

2.2.4. Direct application of SCGs

In this method, SCGs are applied directly onto agricultural land. The emissions to air and leachates from using SCGs as fertiliser as well as the credits for avoiding the production of chemical fertilisers follow the same approach as for composting (see the previous section). For the emissions from the application of SCGs to land, see Table S4 in the SI.

2.2.5. Incineration with energy recovery

Both CHP and electricity-only incineration plants are considered. Taking UK conditions as the basis, their respective share is 20% and 80% (DEFRA, 2013; Nixon et al., 2013). Therefore, the inventory data in Table 4 represent the weighted average taking this share into account. The gross electricity efficiency of both types of plant is assumed at 25% and thermal efficiency at 6.5% (Defra, 2013).

The electricity and heat produced from incinerating SCGs and their corresponding emissions to the environment have been estimated following the method proposed by Doka (2009) and using the Ecoinvent tool for modelling incineration of municipal solid waste (MSW) (Ecoinvent, 2008). However, the modelling has been carried out for the specific composition of SCGs (Table S3 in the SI). It has been assumed that the incinerator consumes 7% of electricity generated (US EPA, 2014) and that thermal distribution losses amount to 5% (DEFRA, 2013).

2.2.6. Landfilling with energy recovery

In the European Union, most of the landfilling facilities recover biogas (EEA, 2017). The energy capacity of the biogas produced (2563MJ/t SCGs) has been estimated using the Ecoinvent tool for sanitary landfills (Ecoinvent, 2008), specifying the composition of SGCs (Table S3). The use of biogas has been modelled according to EEA (2017), which estimates that 30% of landfill gas (769MJ/t MSW) is vented to the atmosphere. From the remaining biogas, 59% is used for electricity production (1333MJ/ t SCG) and 11% is flared (282MJ/t SCG). For electricity generation, a spark ignition engine has been considered, assuming a 38% efficiency (EA, 2010) and internal electricity consumption of 7% (US EPA, 2018). Thus, the total electricity exported to the grid is estimated at 471MJ/t SCGs. The inventory data are summarised in Table 4.

2.2.7. Scenario analysis

To evaluate the potential environmental benefits of a higher value-added valorisation route, i.e. production of biodiesel, four scenarios have been considered, based on the total amount of SCGs produced in the UK annually (256.8 kt of SCGs, Table 5). The scenarios consider the replacement of the predominant current management practices – landfilling and incineration – with biodiesel in different proportions, and also a hypothetical case where all SCGs are used to produce the biofuel. The scenarios are compared to the current SCG treatment practices in the UK, defined as 'business as usual' (BAU), also shown in Table 5; for further details on the current treatment, see Figure S1 and the accompanying text in the SI.

2.3. Impact assessment

GaBi 8.7 software (Thinkstep, 2018) has been used to model the different SCGs valorisation routes. The Recipe 2016 (V1.1) impact assessment method (Huijbregts et al., 2017) has been applied to calculate the environmental impacts, according to the hierarchist perspective. The ReCiPe method has been selected as it represents the state-of-the-art in impact assessment methods and is widely used in LCA studies. It provides a wide set of categories, allowing to consider impacts to air, water, soil, human and ecological health. The characterisation factors are relevant to the European context and hence appropriate for this study.

All 16 impact categories included in Recipe are considered, as follows: climate change (CC), fossil depletion (FD), metal depletion (MD), fine particulate matter formation (PM), stratospheric ozone depletion (OD), photochemical oxidant - ecosystems (POFe), photochemical oxidant - humans (POFh), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial acidification (TA) human toxicity, cancer

Table 5
Scenario analysis.

Scenario	Description	Incineration	Landfilling	Anaerobic digestion	Composting	Direct application	Biodiesel
BAU	Current SCGs management practices in the UK	45%	30%	6%	4%	15%	0%
SC1	As BAU but replacing landfilling with biodiesel	45%	0%	6%	4%	15%	30%
SC2	As BAU but replacing incineration with biodiesel	0%	30%	6%	4%	15%	45%
SC3	As BAU but replacing both landfilling and incineration with biodiesel	0%	0%	6%	4%	15%	75%
SC4	All SCGs are used for biodiesel production	0%	0%	0%	0%	0%	100%

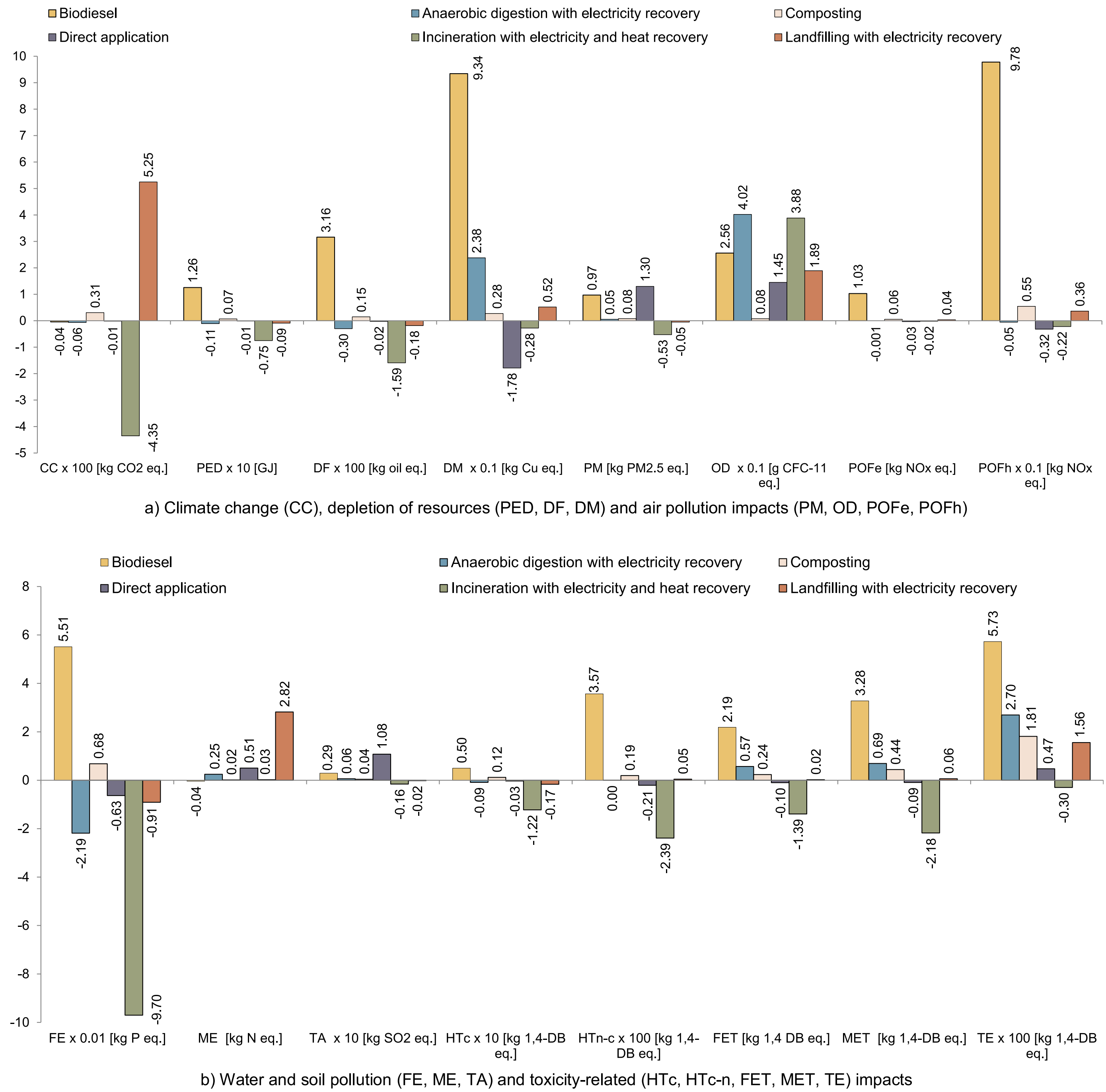


Fig. 2. Comparison of environmental impacts of current waste management practices and biodiesel production from spent coffee grounds (SCGs) [Values expressed per functional unit of 1 t of SCGs. Incineration impacts represent aggregated impacts of heat & electricity and electricity-only incinerators weighted in a proportion of 20%:80%. Some impacts have been scaled and should be multiplied by the factor shown on the x-axis to obtain the original values. CC: climate change; PED: primary energy demand; FD: fossil depletion; MD: metal depletion; PM: fine particulate matter formation; OD: stratospheric ozone depletion; POFe: photochemical oxidant ecosystems; POFh: photochemical oxidant humans; FE: freshwater eutrophication; ME: marine eutrophication; TA: terrestrial acidification; HTc: human toxicity, cancer; HTn-c: human toxicity, non-cancer; FET: freshwater ecotoxicity; MET: marine ecotoxicity; TE: terrestrial ecotoxicity; DCB: dichlorobenzene].

(HTc), human toxicity, non-cancer (HTn-c), freshwater ecotoxicity (FET), marine ecotoxicity (MET) and terrestrial ecotoxicity (TE). In addition to these, primary energy demand (PED) has also been calculated, following the GaBi method (Thinkstep, 2018). Biogenic carbon storage in SCGs is not considered but biogenic methane generated during processing or application of SCGs is included.

3. Results

This section first compares the environmental impacts of the six SCGs valorisation methods considered in the study. This is followed in Section 3.2 by the scenario analysis at the UK level which evaluates the impacts of the differing shares of these routes in an overall SCG management system. Finally, Section 3.3 explores whether following different waste valorisation hierarchies, driven by the circular economy and bio-economy strategies, lead to more sustainable outcomes. All the results are presented per functional unit (treatment of 1 tonne of SCGs).

3.1. Comparison of SCGs valorisation routes

Compared to the other SCGs valorisation routes, biodiesel production is one of the least environmentally sustainable options. As illustrated in Fig. 2, it has the highest impacts in 11 out of 16 categories, including depletion of resources (PED, DF and DM), air pollution (POFe and POFh), FE and toxicity-related impacts (FET, MET, HTn, HTn-c and TE). This is due to the methanol production process: whilst recovering 43% of methanol reduces these impacts, this is insufficient to compete with the energy recovery options, i.e. anaerobic digestion, incineration and landfilling. The latter options benefit from the credits for displacing fossil-fuel-dominated UK electricity and heat and also for displacing chemical fertilisers in the case of anaerobic digestion. As a result, incineration is the most environmentally sustainable valorisation route, with net-negative impacts in 14 out of 16 categories, including climate change (CC). Therefore, decarbonisation of the electricity mix and chemical feedstocks, in this case methanol, will be key for improving the comparative environmental performance of the emerging SCG valorisation routes, such as biodiesel production.

The environmental impacts of the valorisation routes are discussed in more detail in the next sections, referring to the results shown in Fig. 2; for the contribution of different life cycle stages to each route, see Figs. S2–S8 in the SI. It should be noted that the results for incineration discussed below represent the aggregated impacts of the heat & electricity and electricity-only incinerators, weighted according to their aforementioned share in the UK of 20% and 80%, respectively; the environmental impacts of each type of incineration system can be found in Figure S6 in the SI. All the impacts discussed below are expressed per tonne of SCGs treated.

3.1.1. Climate change (CC)

Four out of the six valorisation routes have a net-negative CC impact, meaning that they save carbon emissions, largely due to the credits for recovering useful products. Incineration is the best option, saving 435.3 kg CO_2 eq. Anaerobic digestion is the next best alternative with −6.2 kg CO_2 eq., followed by biodiesel at −4.3 kg CO_2 eq. and direct application of SCGs with −1.3 kg CO_2 eq. Composting and landfilling are net-positive with respect to CO_2 eq. emissions, with 30.7 and 524.7 kg CO_2 eq., respectively. The credit for electricity generation is the dominant factor for biodiesel, anaerobic digestion, incineration and landfilling (see Figures S2, S3, S7 and S8). Anaerobic digestion, composting and direct application also benefit from the credits for avoiding chemical fertilisers, in particular due to the avoidance of N_2O emissions in their production (Figs. S3, S4 and S5). However, N_2O emissions are a key contributor to CC of these three routes from the respective application of the digestate, compost and SCGs to agricultural land. For composting, CO_2 emissions associated with the production of electricity used in the conditioning and composting processes (forced aeration and irrigation) are responsible for a relatively high CC. The landfilling system has the highest impact due to the venting of 30% of biogas (Figure S8) as mentioned in Section 2.2.6.

In the case of biodiesel, system credits reduce CC from 402.5 kg to −4.3 kg CO_2 eq. (Fig. S2). The largest reductions are due to the electricity exported to the grid from the defatted SCGs (−170 kg CO_2 eq.) and the avoided CO_2 emissions from using biodiesel instead of diesel (−188.5 kg CO_2 eq.). Although recovering methanol reduces the demand for the virgin feedstock by nearly a half (43%), the impact is still driven by CO_2 and CH_4 emissions associated with steam methane reforming of methanol. However, the analysis carried out as part of this work suggests that replacing this process with methanol from biomass would reduce CC by up to 41 times, to −176.85 kg CO_2 eq. (see Figure S9). Additionally, other four impacts (DF, PM, OD & TA) would decrease on average by ~15 times, with DF being net-negative. However, using methanol from biomass also increases the other 11 impacts; ranging from 36% higher POFh to 6.7 times greater ME.

Therefore, the gains in climate change and a small number of other impacts would be achieved at the expense of the vast majority of other impacts.

There are no other studies of SGC biodiesel produced by the one-step the esterification process considered here. The only other study available on SCGs biodiesel at the time of writing is that by Kokoos (2018) who considered a two-step esterification process. However, the system boundary was from cradle to gate, considering only the production process and excluding other life cycle stage. If the CC impact obtained in the current study is recalculated for the same cradle-to-gate boundary, it is 32 times higher than in Kokoos: –0.065 vs –2.1 kg CO_2 eq./kg biodiesel (both values including biogenic carbon storage). The main reason for this is the difference in the two production processes, including different types and quantities of solvents.

In comparison to other biodiesel fuels produced from waste, the impact estimated here is within the range: –1.65 g CO_2 eq./MJ,[1] compared to –88 to 80 g CO_2 eq./MJ (RAEng 2017). Relative to fossil diesel (83.8 g CO_2 eq./MJ (EC, 2015)), it reduces the carbon emissions well below the 60% required by the EU Renewable Energy Directive (EC, 2015) for new production plants.

3.1.2. Resource depletion (PED, DF, DM)

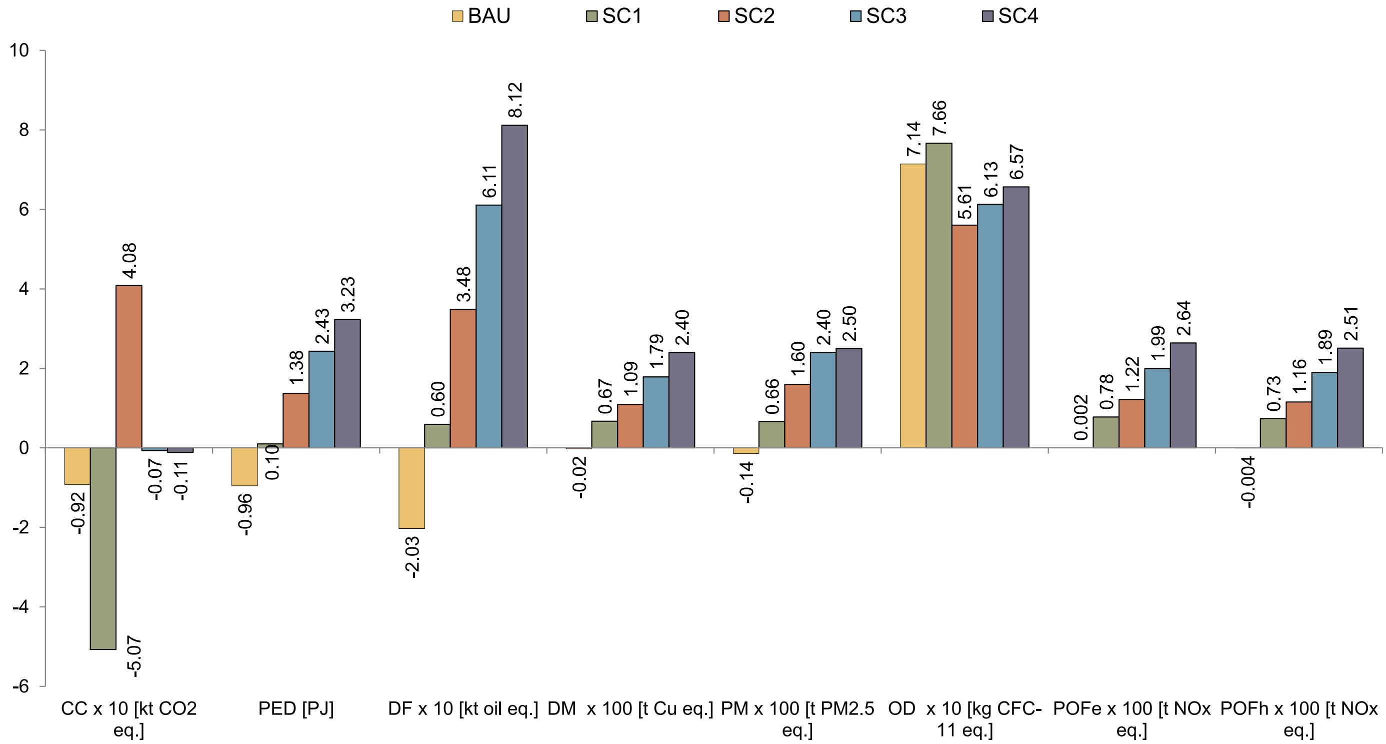
As can be seen in Fig. 2a, biodiesel has the highest values for all the resource-related impacts, followed by composting. The energy credits lead to net-negative PED and DF for incineration (–7.5 GJ and −159 kg oil eq.), landfilling (–894.8 MJ and –18.4 kg oil eq.) and anaerobic digestion (–1.1 GJ and –29.9 kg oil eq.). Comparable effects are seen due to the credits for avoiding chemical fertilisers for direct SGC application (–109 MJ and –2.5 kg oil eq.) and for the aforementioned impacts from anaerobic digestion. The PED and DF of composting are estimated at 677 MJ and 14.8 kg oil eq., respectively. Similar to CC, the methanol required in the production of biodiesel is nearly the only source of PED and DF, which is mainly driven by the natural gas used in the steam reforming process (~96%).

Only two valorisation routes exhibit net-negative DM: direct SGC application (–178.4 g Cu eq.) and incineration (–27.6 g Cu eq.). Biodiesel has the highest impact (934 g Cu eq.), followed by anaerobic digestion (237.9 g Cu eq.), landfilling (51. g Cu eq.) and composting (27.6 g Cu eq.). The use of metals, in particular iron and nickel in the facilities, plants and machinery is the main source of DM for all the valorisation routes. Additionally, the use of copper and molybdenum in the life cycle of methanol is also significant for the impact from the biodiesel system. Although credits from electricity and heat generation partly offset DM, in particular for incineration and landfilling, credits for the avoidance of chemical fertilisers play a larger role, especially due to the avoidance of phosphorus. This is particularly important for direct SCG application and anaerobic digestion (for details, see Figs. S2–S8 in the SI).

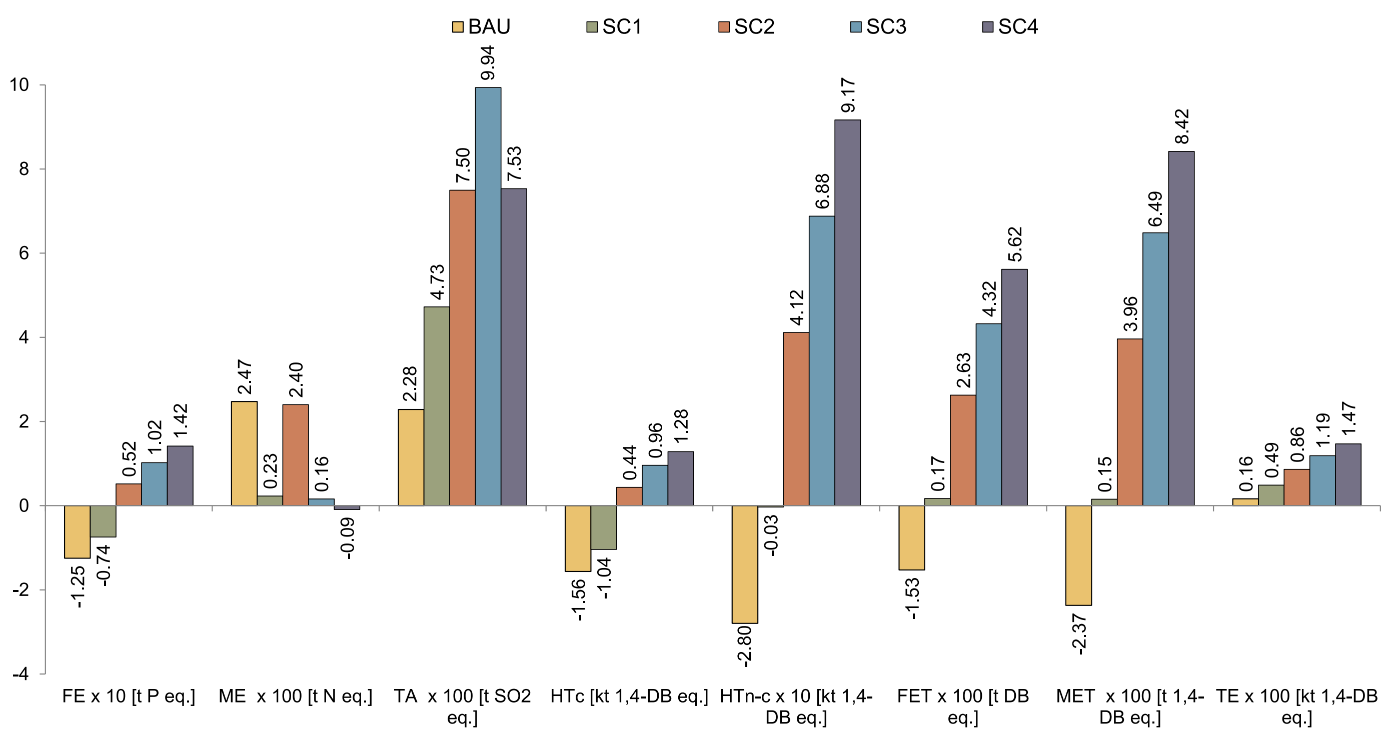
3.1.3. Air pollution (PM, OD, POFe, POFh)

Credits associated with electricity and heat recovery, mainly due to the avoidance of SO_2 emissions, help to counteract the formation of PM related to incineration and landfilling. As a result, these technologies have a net-negative impact (–527 and –50 g $PM_{2.5}$ eq., respectively). Although the credits reduce PM across all the valorisation routes, NH_3 emissions related to the use of SCG as N-based fertiliser are the main source of impact from anaerobic digestion due to the digestate (54 g $PM_{2.5}$ eq.) and compost (81 g $PM_{2.5}$ eq.) as well as direct application of SCGs (1.3 kg $PM_{2.5}$ eq.). SO_2 emissions from methanol production are the main contributor to the high PM associated with biodiesel (973 g $PM_{2.5}$ eq.).

[1] Biodiesel heating value is 39.6 MJ/kg (Patra et al., 2016) and the yield is 66.15 g biodiesel/kg SCGs (see Table 1).



a) Climate change (CC), depletion of resources (PED, DF, DM) and air pollution (PM, OD, POFe, POFh) impacts



b) Water and soil pollution (FE, ME, TA) and toxicity-related (HTc, HTc-n, FET, MET, TE) impacts

Fig. 3. Comparison of the environmental impacts of the current SCG treatment practices at the UK level and potential scenarios for replacing incineration and landfilling with biodiesel production. [All impacts expressed per year. For scenario descriptions, see Table 5. For the nomenclature, see Fig. 2. Some impacts have been scaled and should be multiplied by the factor shown on the x-axis to obtain the original values.].

As seen in Fig. 2a, OD is one of the few impacts for which biodiesel (0.26 g CFC-11 eq.) is more competitive against incineration (0.39 g CFC-11 eq.) and anaerobic digestion (0.4 g CFC-11 eq.). This is due to the smaller benefits from the energy-related credits for this impact. N_2O emissions are the main cause of OD across all the valorisation routes. In the case of biodiesel, the avoidance of diesel and glycerine production helps to reduce this impact. Similarly, the credits for avoiding the production of N-based chemical fertilisers are also important for anaerobic digestion, composting (0.8 mg CFC-11 eq.) and direct SGC application (0.15 g CFC-11 eq.).

Biodiesel is the worst option for POFe and POFh (1 and 0.96 kg NO_x eq.) while direct SGC application is the best, exhibiting net-negative impacts (–31 and –32 g NO_x eq.). Incineration (–18 & –22 g NO_x eq.) and anaerobic digestion (–1 & –5 g NO_x eq.) also have net-negative impacts. Emissions of NO_x and non-methane volatile organic compounds (NMVOC) are the main contributors in all the routes, except for incineration and landfilling, where N_2O from combustion also influences these impact categories.

3.1.4. Water and soil pollution (FE, ME, TA)

As illustrated in Fig. 2b, four routes have net-negative FE: incineration (–97 g P eq.), anaerobic digestion (–22 g P eq.), landfilling (–9 g P eq.) and direct SCG application (–6 g P eq.). The credits for the avoidance of PO_4 emissions from energy and chemical fertilisers are the main reasons for the savings in this impact. By contrast, biodiesel and composting exhibit net-positive FE, with 55 and 7 g P eq., respectively. PO_4 emissions from methanol production and electricity generation are the main sources of this impact.

Of the three impacts considered in this section, ME is the only category for which biodiesel shows the lowest value and is net-negative (–36 g N eq.). This is due to the avoidance of NO_3^- emissions related to the credits for glycerine production. On the other hand, NO_3^- emissions for using SCG as N-fertiliser are the core reason for the high impact from direct SCG application (505 g N eq.), anaerobic digestion (247 g N eq.) and composting (15 g N eq.). Despite the credits for energy recovery, NH_3 and NO_3^-emissions make landfilling the worst route, with 2.8 kg N eq. The impact from incineration is estimated at 25 g N eq.

Similar to ME, the N-emissions from using SCGs as fertiliser, especially NH_3, drive TA. The latter is the highest for direct SGC application (10.8 kg SO_2 eq.), while anaerobic digestion (0.6 kg SO_2 eq.) and composting (0.4 kg SO_2 eq.) have a lower impact. The avoidance of SO_2 emissions associated with combustion of fossil fuels accounts for the net-negative TA of incineration (–1.6 kg SO_2 eq.) and landfilling (–0.18 kg SO_2 eq.). However, the equivalent credits for biodiesel are not sufficient to lead to a net-negative impact (2.9 kg SO_2 eq.). SO_2 emissions from the life cycle of methanol production are the main reason for the high TA of biodiesel, positioning this valorisation route as the worst option.

3.1.5. Toxicity-related impacts (HTc, HTc-n, FET, MET, TE)

Incineration has the lowest and biodiesel the highest values for all the toxicity-related impacts. Human toxicity is driven by water emissions of chromium (HTc), zinc and arsenic (HTn-c) from the construction of the facilities and machinery, as well as from electricity generation. Hence, the credits for energy recovery and the avoidance of chemical fertilisers are critical, leading to the net-negative HTP impacts (and FET) for incineration and direct SCG application. Landfilling and anaerobic digestion also have net-negative HTc (–1.7 and –0.87 kg 1,4-DB eq., respectively); anaerobic digestion also shows a negative value in HTn-c (– 0.34 kg 1,4-DB eq.). The high electricity consumption and a smaller displacement of chemical fertilisers render composting the second least preferable option, after biodiesel.

Emissions of copper and zinc to water are the main causes of FET and MET. For biodiesel, in addition to the aforementioned contributors, emissions of silver and barium from the life cycle of methanol are also relevant. Credits from electricity generation largely contribute to the avoidance of nickel and zinc emissions, reducing biodiesel's FET and MET to 2.2 and 3.2 kg 1,4-DB eq., respectively. The electricity credits are the main drivers of the net-negative MET and TE for incineration (–238.8 and –29.8 kg 1.4-DB eq., respectively). Similarly, chemical fertilisers replaced by direct SCG application are the sole reason for the net-negative FET and MET (–0.1 and 0.09 1.4-DB eq., respectively). Overall, TE is the only impact where transport has an important contribution across all the valorisation routes, in particular for anaerobic digestion (269.6 kg 1.4-DB eq.) and composting (181.2 kg 1.4-DB eq.). Emissions of heavy metals to air, mainly copper and antimony, are the key contributors to this impact category.

3.2. Scenario analysis

As indicated in Fig. 3, taking into account the annual amount of SCGs in the UK, the current mix of management options (BAU) has the lowest impacts in 13 out of 16 categories. Of these, 11 categories are net-negative, including CC and depletion of resources (PED, DF and DM).

However, the scenario where landfilling is replaced by biodiesel production (SC1) is the best option for CC at –50.7 kt CO_2 eq./yr. This is 5.5 times lower than the value for BAU of –9.2 kt CO_2 eq./yr. Although scenarios SC3 and SC4 also have net-negative values (–0.69 and –1.1 kt CO_2 eq./yr), they are not competitive enough against the BAU. Replacing incineration with biodiesel (SC2) is the worst alternative for CC (40.8 kt CO_2 eq./yr). Contrary to CC, SC2 has the lowest OD (56.1 kg CFC-11 eq.) since biodiesel has a lower impact than incineration and landfilling (see Fig. 2a). This is also seen in the reduction of OD found in SC3 and SC4 when compared to BAU (61.3 and 65.7 vs 71.4 kg CFC-11 eq.)

Compared to BAU, using all the SCGs for biodiesel (SC4) has much higher impacts than BAU in 13 categories. The greatest increase is found for POFh, which is 1061 times greater for SC4, and for DM, which is 50 times higher than BAU.

Overall, production of biodiesel from SCGs is not yet environmentally a competitive option, in particular when replacing incineration (SC2-SC4), as almost all impacts (13 out of 16) increase. The replacement of landfilling SCGs to produce biodiesel (SC1) is the most competitive option, showing net-negative impacts in four categories (CC, FE, HTc and HTn-c). Only three impacts (CC, OD and ME) decrease on the BAU levels when replacing incineration with biodiesel (SC2-SC4). In the case of OD, the replacement of incineration by biodiesel (SC2-SC4) reduces relative to BAU by up to 22% (SC2); the opposite trend is seen when biodiesel replaces landfilling (SC1) as the impact increases by 7%. Finally, along with CC (–1.1 kt CO_2 eq.), ME is the only other impact for which the full replacement of the current options by biodiesel (SC4) has a net-negative value (–9.3 t N eq.).

3.3. Waste valorisation hierarchy and environmental impacts

The SCGs valorisation routes are classified in Fig. 4 according to the waste hierarchy guidelines (EC, 2008; DEFRA, 2011). The guidelines suggest the most and least preferable options for managing waste, aiming to reduce environmental impacts and increase resource efficiency (EC, 2017a). This has been set in Article 4 of the revised Waste Framework (Directive 2008/98/EC) (EC, 2008) and is considered a crucial guide for the future transition to a circular economy (EC, 2017a). As shown in Fig. 4, 'preventing waste' is the most preferable option in the waste hierarchy; when waste is unavoidable, 'prepare for re-use' is the next best alternative to keep the products (resources) for longer within the system. The third option is 'recycling' to convert waste into new products or materials. This is followed by 'other recovery', which refers to recovery of energy and materials from waste (EC, 2012). Finally, disposal (landfilling) is the least preferable option.

To help with the implementation of a circular economy, along with more sustainable production and consumption, a waste-to-energy process guideline has been developed to complement the aforementioned waste hierarchy (EC, 2017a). This guideline aids positioning of waste-to-energy technologies across the waste management preferences described in the waste hierarchy. For instance, for recycling, anaerobic digestion is considered the most desirable waste-to-energy alternative, followed by incineration with high-energy recovery and the use of

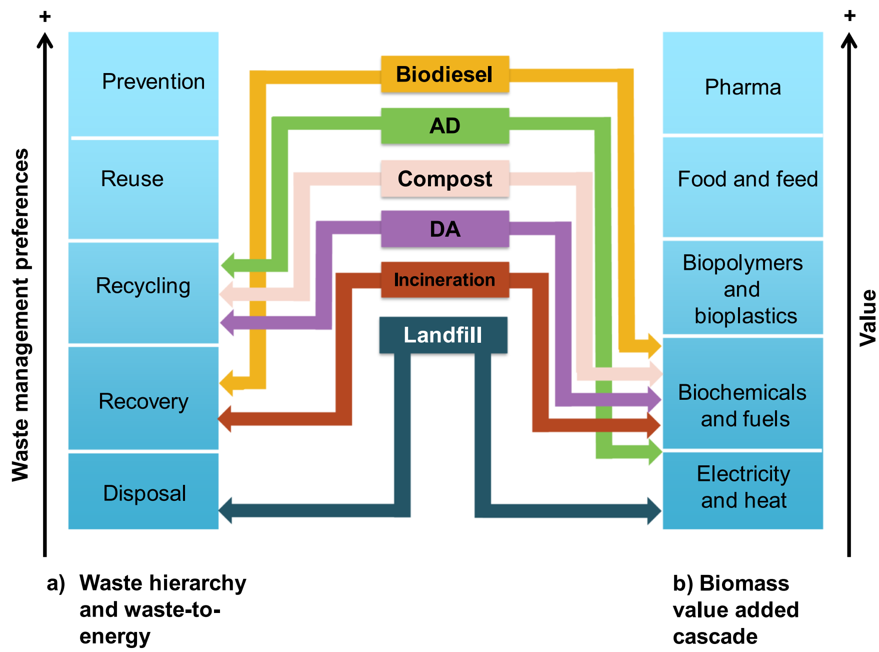


Fig. 4. Classification of spent coffee grounds valorisation routes considered in this study. [Left side of the figure: waste hierarchy and waste-to-energy technologies based on the EC (2008) and EC (2017) guidelines. Right side: biomass value added cascade (BVC) for bio-based products (Lange et al., 2012).].

waste to produce solid, gaseous and liquid fuels, as part of the 'other recovery' option (Fig. 4). The least desirable waste-to-energy processes are incineration with low-energy recovery and landfilling with biogas capture (EC, 2017a). Finally, the "biomass value cascade" (BVC) (Lange et al., 2012) has also been proposed to evaluate the value of bio-resources recovered from waste. This hierarchy prioritises high value-added products from a valorisation route. Consequently, pharmaceutical products are ranked as most desirable, followed by food and animal feed. Middle-ranging products are bio-polymers and bio-plastics, followed by bio-fuels and bio-chemicals. Finally, the lowest value-added products are electricity and heat.

As illustrated in Fig. 4, the SCGs valorisation routes evaluated in this work belong to the bottom three options in the waste hierarchy – recycling, recovery and disposal (EC 2008). Similarly, when considering value-added products according to BVC, recovering energy and heat from waste, as most of the current SCGs treatments options do, is classified as a low-value route. Biodiesel can be categorised as a middle-ranking waste valorisation route. The use of SCGs as fertiliser (anaerobic digestion, composting and direct application) can be classified within BVC as a bio-chemical option because they replace chemical fertilisers. However, it is more difficult to classify composting and direct application of SCG within the waste hierarchy. In this study, these are considered as part of recycling; however, reuse could also be an option (EC, 2012).

As seen in Fig. 5, the ranking of the SGC valorisation options differs between the three waste valorisation hierarchies discussed above and the estimated environmental impacts, all assumed here to have equal importance. Owing to the high energy content in SCGs and a high contribution of fossil fuels to grid electricity in the UK, incineration with electricity recovery is the best option based on the environmental impacts, in particular for climate change and the resource-related impacts (Fig. 5). This is in contradiction to all three waste valorisation hierarchies where incineration and energy recovery are the least preferred options. Similarly, composting, widely practised in food waste management, exhibits a poor environmental performance, with impacts higher than incineration, anaerobic digestion, direct SCG application and even landfilling. The only two options which rank similarly for both the valorisation hierarchies and the environmental impacts are direct SGC application and anaerobic digestion. Interestingly, biodiesel production, which ranks higher in BVC, has the highest environmental impacts.

Despite the aforementioned guidelines being flexible, this is an example of how difficult it is to select and prioritise valorisation routes, particularly without quantitative information on their environmental impacts. Therefore, qualitative waste valorisation guidelines should always be supported by quantitative environmental assessments based on LCA. This is particularly important if government incentives are to be introduced to promote the commercialisation of emerging biotechnologies, as expected in the UK in connection with the its bioeconomy strategy (Vanderhoven and Corbett, 2018; HM Government, 2015). Additionally, it is imperative to set priorities in terms of resource scarcity and decarbonisation of energy and feedstocks, specifying national targets, to help address the trade-offs between waste-to-energy processes and increasing the valued-added of waste (bio-products).

Along with this, the credits for energy recovery are critical as they can affect the environmental impacts significantly. If the grid is decarbonised, the credits will be lower, hence the energy recovery options, such as incineration and anaerobic digestion, may not be as environmentally sustainable as they appear at present. On the other hand, a greater contribution of renewables on the grid will lead to greater depletion of metals (Stamford and Azapagic, 2014), increasing the credits for this impact.

Furthermore, the definition of the functional unit will also influence the results, as seen in this study; when the functional unit is related to waste treatment, biodiesel does not perform well enough to compete with the current routes. However, when the functional unit is based on energy content (MJ), it does show benefits relative to diesel and petrol (see Section 3.1.1). Other functional units can also be considered, including the amount of energy or materials recovered. Therefore, future work should explore a number of functional units, congruent with the related goal and scope of the study, to evaluate the effect on the results.

4. Conclusions

This study has evaluated the environmental impacts of six SGC valorisation routes. The most environmentally sustainable option is incineration, with 14 net-negative impacts out of 16 considered in the study.

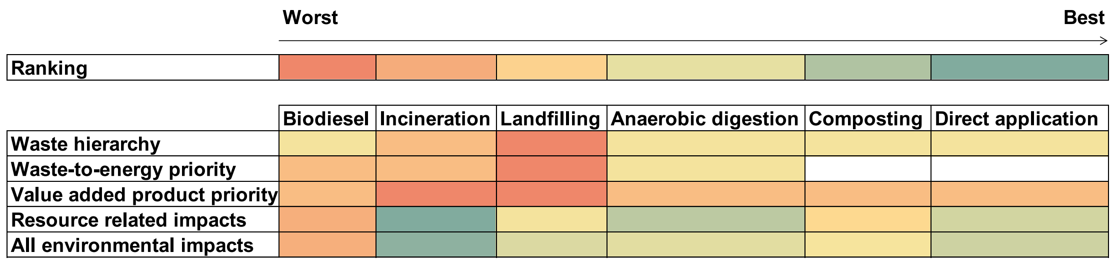


Fig. 5. Comparison of valorisation routes based on the waste hierarchy (EC, 2008), waste-to-energy process (EC, 2017a), biomass value cascade (value added) (Lange et al., 2012) and the estimated aggregated environmental impact. [*Colour coding corresponds to the scale of* 1 (*red* = *worst option*) *to* 6 (*green* = *best option*). *The full scale is used for* "Resource related impacts" *and* "All environmental impacts". *For* "Waste hierarchy" *a scale of* 1–3 *is used as there are only three options based on the hierarchy in* Fig. 4 (*left-hand side*): *disposal* (*landfilling*) = 1, *recovery* (*incineration*) = 2 *and recycling* (*anaerobic digestion, composting and direct application*) = 3. *Similar applies to* "Waste-to-energy priorities": *landfilling* = 1, *incineration* = 2 *and anaerobic digestion* = 3; *composting and direct application do not fit in this category* (*blank*). *The* "Valued added product priority" *is ranked from* 1 *to* 2 *as there are only two options* (Fig. 4, *right-hand side*): *electricity and heat* (*incineration and landfilling*) = 1, *bio-chemicals and fuels* (*anaerobic digestion, composting, direct application and biodiesel*) = 2. "Resource related impacts" *comprise primary energy demand, climate change, depletion of fossil resources and metals*. "Resource related impacts" *and* "All environmental impacts" *are aggregated assuming equal preference for each of the categories considered. For the latter, see* Figure S10 *in the* SI].

This is followed by direct application of SCGs as fertiliser with 11 net-negative categories. Anaerobic digestion and landfilling are mid-ranking routes, with eight and six impacts being net-negative, respectively. Finally, although biodiesel production has net-negative climate change and marine eutrophication, it is the least preferred option with the highest impacts in 11 categories, followed by composting. When considering possible scenarios for SCGs management within an integrated system, the introduction of biodiesel is only competitive for climate change and marine eutrophication, and only when replacing landfilling.

While it is expected that biodiesel production would contribute towards climate change mitigation and more efficient use of resources, the life cycle assessment shows a different perspective. The high energy generated by waste-to-energy processes and credits for displacing the fossil-fuel dominated electricity mix reduce the impacts of these valorisation routes. In addition, the high consumption of fossil-derived methanol in biodiesel production is the main reason for its poor environmental performance, even with nearly half of methanol recovered. If methanol produced from biomass is used instead, climate change and other four impacts are reduced, but other 11 impacts increase.

Hence, it is clear that, to promote the development of bio-technology options, efforts towards decarbonisation of energy, in particular electricity, and feedstocks such as methanol, are critical. Furthermore, in terms of using LCA in decision-making, it is important to standardise the methodology, including the functional unit(s) and system boundaries, to ensure consistent and robust analyses and decisions. It is recommended that for policy-making, LCA studies should include multiple functional units, to explore the effect on the findings. It is also recommended to evaluate the economic and social implications of the options under analysis, to provide a comprehensive set of decision indicators and aid the selection of the most sustainable alternative.

Finally, the results also show how the ranking of different options differs when considering life cycle environmental impacts from the ranking according to different waste valorisation hierarchies. This necessitates the need for harmonisation of different waste valorisation hierarchies and their integration with quantitative life cycle analysis to ensure development of a sustainable circular economy.

CRediT authorship contribution statement

Ximena C. Schmidt Rivera: Conceptualization, Methodology, Formal analysis, Visualization, Writing - original draft. **Alejandro Gallego-Schmid:** Conceptualization, Methodology, Formal analysis, Visualization, Writing - original draft. **Vesna Najdanovic-Visak:** Data curation, Writing - review & editing. **Adisa Azapagic:** Funding acquisition, Methodology, Project administration, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare no conflict of interest.

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Supplementary materials

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