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Effects of system design and Co-product treatment strategies on the life cycle performance of biofuels from microalgae

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A B S T R A C T

This study presents a life cycle greenhouse gas and energy assessment for two algal biofuel production pathways: biodiesel produced through lipid extraction (LE) and renewable diesel produced through hydrothermal liquefaction (HTL). The two production pathways generate different co-products, which are handled through allocation in life cycle assessment-based analyses. The method and assumptions used for co-product allocation affect the performance of the analyzed fuels, and are thus examined through scenario analysis; five co-product allocation strategies are tested for the LE pathway and six are tested for the HTL pathway. After allocation, the carbon intensity of renewable diesel varies from 36 to 54 gCO_2e/MJ, and the primary energy consumption of renewable diesel varies from 0.7 to 1.2 MJ/MJ; while the carbon intensity of biodiesel ranges, remarkably, from −59 to 125 gCO_2e/MJ, and the primary energy consumption of biodiesel ranges from 0.1 to 1.7 MJ/MJ. The optimal algal oil production pathway is determined by comparing open-loop and closed-loop systems, considering not only the estimated net environmental impacts, but also the confidence or uncertainty of those outcomes.

1. Introduction

Interest in biofuels derived from microalgae as an alternative to traditional energy crops is growing because it may avoid some of the consequential effects of terrestrial oil crops (Faried et al., 2017). However, microalgae require a large amount of fertilizer during cultivation to achieve high oil productivity. And the energy input during harvesting and dewatering of the biomass is intensive. Many life cycle assessment (LCA) studies of algal oil production have been done to evaluate environmental impacts and identify energy intensive processes of the system. The GHG emissions vary from 20 to 500 g CO_2e/MJ, while the energy return on energy investment (EROI) of microalgae biodiesel ranges from 0.2 to 6 (Quinn and Davis, 2015; Shimako et al., 2016). This range of values is the result of both method- and model-induced variability and real variability in the performance of current and simulated future systems (Raheem et al., 2018; Zaimes and Khanna, 2013; Kendall and Yuan, 2013). The selection of conversion technologies is identified as a major model-induced variable in existing LCAs for microalgae biofuel (Collet et al., 2015; Frank et al., 2013). Among distinct microalgae biofuel conversion pathways, two main pathways that have been discussed the most are renewable diesel production from hydrothermal liquefaction (HTL) and biodiesel production from a solvent-based lipid extraction (LE) process (Davis et al., 2018; Laurens et al., 2017; Tian et al., 2017).

The sources of method-induced variability are many, and among them the methods used to treat co-products stand out as requiring additional study and guidance (Gnansounou and Kenthorai Raman, 2016; Zaimes and Khanna, 2014), especially since LCA has been called on to assist policy making processes to identify the environmental effects of biofuels (Soratana et al., 2014). Most biofuel production processes are multi-functional systems that produce biofuel products along with economically valuable co-products, such as algal biomass residual (algal cake) that may be used as animal feed and fertilizers. Instead of assigning environmental burdens solely to the biofuel, some methods are required to distribute burdens among a biofuel and its co-products.

Allocation methods include partitioning methods and displacement methods. Partitioning methods allocate burdens among products on the basis of a physical or economic value (e.g. energy content, mass or economic value), while displacement expands the analysis to include the displacement effects of a co-

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product on substitutable products in the market (ISO14044, 2006). The displacement method and economic allocation are more frequently recommended than energy and mass-based allocation methods (Lardon et al., 2009; Wang et al., 2011). Cai et al. (2018) conclude that partitioning methods can be subjective; and the displacement method is recommended for co-products with distinct properties. However, the displacement method requires detailed production data of the displacement and its market demand. It is important to examine the co-product's market price, market capacity and the products to be displaced.

An alternative to utilizing co-products in the market is the reuse and recycling of co-products within the production system to reduce material inputs. This leads to a closed-loop production system. A closed-loop system avoids uncertainties from co-product allocation issues and is advocated under the concept of circular economy (Murray et al., 2017). The allocation methods used for partitioning environmental burdens to primary products such as biofuels and co-products and the assumption of how co-products are utilized can significantly affect the results of an LCA (Hoefnagels et al., 2010). Different allocation methods might shift the energy balance and carbon reduction for biofuel from negative to positive (Zaimes and Khanna, 2014), indicating the potential of arbitrary allocation decisions affected by different co-product utilization choices.

Numerous studies have tested the weaknesses and advantages of each allocation method, and sometimes a hybrid allocation approach is employed to present a realistic utilization of the energy products and co-products (Canter et al., 2016; Mackenzie et al., 2017). However, in previous microalgae based LCA studies, co-product allocation strategies are mainly discussed for biodiesel production rather than other conversion processes (Gnansounou and Kenthorai Raman, 2016; Zaimes and Khanna, 2014), meaning there has been limited assessment of the effects of co-products for other microalgae biofuel production systems. Thus a comparison of the effect of co-products and allocation method choices between different production systems fills an existing research gap.

In this study, we have considered potential applications of co-products from the two microalgae biofuel production pathways (HTL and LE), and investigated different treatment methods within harmonized system boundaries. As part of this analysis, the market potential of each co-product is evaluated to determine the displacement credit and variability induced by displacement credit.

2. Materials and methodology

2.1. Goal and scope

The objective of this study is to evaluate and compare the life cycle GHG emissions and energy performance of biodiesel and renewable diesel produced from microalgae through two technology pathways under different co-product treatment strategies using a process-based, prospective LCA approach. LCA is a technique for evaluating the environmental aspects and potential environmental impacts of a product throughout its life cycle, considering the full supply chain of inputs (ISO14040, 2006). Life cycle energy and GHG assessments are a narrow application of the LCA method, since full LCA considers a suite of impact categories.

The research presented here applies this narrow form of LCA, accounting for energy, direct water consumption (meaning indirect and upstream water use are not accounted for) and global warming potential (GWP). Energy and water consumption are reported simply as inventory values (e.g. MJ of energy and liters of water). GHGs are reported in units of CO_2-equivalent (CO_2e). The IPCC's 100-year GWPs are used to convert non-CO_2 emissions into CO_2e (28 for biogenic CH_4, 30 for fossil CH_4, and 265 for N_2O) (IPCC, 2013). This means that 1 kg of methane released is equivalent to 30 kg of CO_2 released when assessed over a 100 year period.

2.2. System definition and boundary

The system boundary of the two pathways (the LE pathway and HTL pathway) is illustrated in Fig. 1. Biodiesel is produced from the LE pathway, and renewable diesel is produced from the HTL

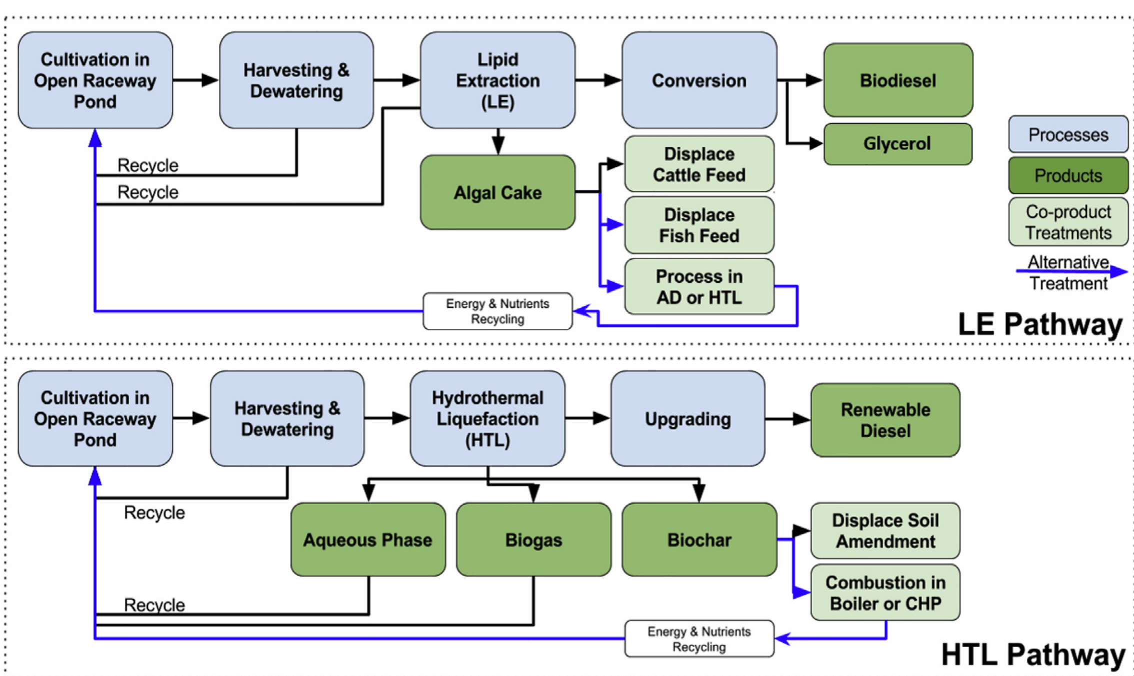

Fig. 1. System description of algal biofuel production through LE and HTL pathway.

Table 1
Growth model assumptions and input summary for cultivation, harvesting and dewatering (all parameters are dry weight based).

Modified Growth Model		
Parameter settings	Unit	Input
Growth rate[a]	g/(m^2•d)	25.00
Lipid content[a]	wt%	25.00
Protein[a]	wt%	32.15
Carbohydrate[a]	wt%	34.85
Ash[a]	wt%	8.00
C[a]	g/kg biomass	500.00
N[a]	g/kg biomass	52.50
P[a]	g/kg biomass	12.92
CO_2 requirement[a]	kg/kg biomass	1.83
CO_2 use efficiency[a]		0.87
Ammonium nitrate (NH_4NO_3) requirement[b]	kg/kg biomass	0.15
Triple superphosphate ($Ca(H_2PO_4)_2$) requirement[b]	kg/kg biomass	0.10
Energy for CO_2 injection[a]	MJ/kg biomass	0.18
Energy for paddlewheel[a]	MJ/kg biomass	0.68
Energy for water pumping[a]	MJ/kg biomass	0.78
Energy for water pumping within the system[a]	MJ/kg biomass	0.76
Mixing energy for flocculation[a]	MJ/kg biomass	0.0032
Energy for DAF[a]	MJ/kg biomass	0.1203
Biomass recovery from harvesting[a]		90%
Biomass recovery from dewatering[a]		96%
Electricity for centrifugation[a]	MJ/kg biomass	0.576
Water content after dewatering[a]	L/kg biomass	5.56
Water Evaporation rate[a]	L/(m^2•d)	5.97
Evaporation Loss[a]	L/kg biomass	238.66
Pond Area[a]	ha	161.87
Annual Biomass Yield[a]	t/(ha•y)	75.00

Data source:
[a] Yuan et al. (2015).
[b] Modeled in this study.

pathway. The scope of this analysis is “cradle-to-gate,” meaning that the analysis stops at the biorefinery gate. Thus, the life cycle stages included in the analysis are microalgae cultivation in open raceway ponds (ORPs), microalgae harvesting and dewatering, bio-crude production via LE or HTL, conversion of bio-crude oil into the final energy product (biodiesel or renewable diesel), and utilization of co-products. The transportation and utilization of biodiesel or renewable diesel are excluded from the system boundary. Fig. 1 describes the steps in each of the considered pathways.

The processes of microalgae cultivation, harvesting and dewatering, drying, oil extraction, and utilization of algal cake occur within the same facility. From there the crude oil is transported to a nearby refinery for conversion to biodiesel or renewable diesel. Construction, repair and maintenance of infrastructure, production of equipment and waste management are excluded from the system boundary. The functional unit of analysis is 1 MJ of microalgal biofuel, although 1 kg of dry biomass is used as a modeling unit of analysis to assess the material and energy consumption in each unit process in the life cycle inventory (LCI) assessment.

2.3. The microalgae cultivation, harvesting and dewatering

The cultivation model of the microalgae *Scenedesmus dimorphus,* grown in ORPs, is adopted from previous work (Yuan et al., 2015). The production facility of 162 ha of open raceway ponds are assumed to be located in southern New Mexico (which determines water quality, groundwater depth for water pumping and evaporation rates), with pond dimensions of 100 m by 10 m and a water depth of 0.3 m CO_2 is assumed to be provided by a co-located power plant and directly injected into ponds, which may underestimate the burdens of CO_2 provision depending on CO_2 source and location (Davis et al., 2018). In previous research Yuan et al. (2015) examined four combinations of technologies for harvesting and dewatering, including bioflocculation followed by dissolved air flotation (DAF) and centrifugation, flocculation with polymer followed by DAF and centrifugation, flocculation with alum followed by DAF and centrifugation, and centrifugation only. The most efficient harvesting and dewatering technology route was found to be bioflocculation following DAF and centrifugation. The bioflocculation (or autoflocculation) was achieved by changing culture conditions or using algae produced biopolymers, and thus required no chemical flocculants. The current model uses this as the default harvesting and dewatering route. The conditions for this route are as follows: the initial biomass concentration is 0.5 g/L dry weight (DW), which increases to 50 g/L DW out of the DAF process, and reaches 180 g/L DW after dewatering with the centrifuge. Table 1 summarizes key parameter assumptions, material inputs, and energy inputs during the microalgae cultivation and harvesting stage.

2.4. Microalgae renewable diesel production through HTL pathway

HTL is a thermochemical process involving the reaction of biomass in water at moderate temperatures (250–400 °C) and high pressure (5–30 MPa) for a certain reaction time with or without the use of a catalyst (Baloch et al., 2018). HTL yields a product typically referred to as bio-crude or bio-oil along with gaseous, aqueous (liquid) phase, and solid phase (char) streams. In order to model the HTL process under different operation conditions, a mathematical kinetic HTL model was employed (Valdez et al., 2014).

2.4.1. HTL modeling

The kinetic HTL model developed by Valdez et al. (2014) estimates product quantities including bio-crude oil, aqueous phase, gas phase and solid phase as a function of the characteristics of the microalgae feedstock. The model provides four operating conditions, 250 °C, 300 °C, 350 °C and 400 °C, with retention times ranging from 1 to 90 min. The HTL product yields reflect the biochemical composition of microalgae and the operating conditions of the HTL system. Unfortunately, this kinetic model is not capable of defining the properties of each product. The simplified results may distort real characterizations of HTL products, thus further investigation in process modeling is desired. The C and N content in each product are estimated from empirical data in the literature (as described in section 2.4.3). Below some of the key features and assumptions beyond the kinetic modeling of the HTL technology pathway are described:

- *HTL Process Model:* The HTL process energy demand is assumed to be equal to the energy needed to heat the medium to operation temperature from ambient temperature at 20 °C (Fortier et al., 2014). A spiral tube heat exchanger is integrated in the system, to re-heat the incoming biomass with the outgoing streams from HTL reactor, assuming 80% of HTL heat can be recovered with 85% efficiency (Delrue et al., 2013). Additional energy is needed to meet process energy demands; grid electricity is used for pumping, and natural gas (NG) is used for the remaining heat demand not met by heat recirculation. NG is assumed to be combusted in a boiler with 85% efficiency.
- *HTL Products Separation.* There is currently no consistent method used for separation of the HTL products (Xiu and Shahbazi, 2012). Various methods including water separation, solvent separation, filtration, vacuum and centrifugation were reported to separate solid and oil under lab conditions (Huang et al., 2013; Zacher et al., 2014). Due to the inconsistency and lack of data for scaled application, the separation process is omitted in this analysis.
- *Bio-crude oil Upgrading.* Bio-crude oil from HTL has high potential for co-processing with petroleum bio-crude oil in conventional refineries to produce renewable transportation fuels such as renewable diesel, which has the identical properties as conventional diesel (Jensen et al., 2016). However, the bio-crude has higher oxygen, nitrogen and sulfur content than conventional bio-crude oil. Because of the high oxygen content, an additional process for removing oxygen from the bio-crude, deoxygenation, is recommended before the co-processing (Xiu and Shahbazi, 2012). We assume bio-crude oil can be co-processed directly with petroleum crude in a refinery (Jones et al., 2014; Jensen et al., 2016). The inputs and energy consumption for upgrading bio-crude oil to renewable diesel are adopted from existing studies as listed in Table 2. Hydrogen (H_2) consumption is assumed 3%wt of bio-crude oil, a conservative value from reported range of 1 wt% to 4 wt% feed oil (Wu and Liu, 2016). Inputs and outputs of HTL pathway are summarized in Table 2.

Table 2
Inputs and Outputs Summary of HTL Pathway at 350 °C for 15 min (dry weight based).

Parameter	Unit	Value
Pumping Electricity[a]	MJ/kg biomass	0.001
HTL Natural Gas (NG) [a]	MJ/kg biomass	2.82
Biocrude Oil[a]	kg/kg biomass	0.42
Gas Phase[a]	kg/kg biomass	0.014
Aqueous Phase[a]	kg/kg biomass	0.485
Solid Phase[a]	kg/kg biomass	0.081
Upgrading Electricity[b]	MJ/kg biomass	0.02
Upgrading H_2[b]	kg/kg biomass	0.01
Upgrading Water[c]	L/kg biomass	0.50
Upgrading Efficiency[d]		85%
Renewable Diesel Yield[a]	MJ/kg biomass	17.15
N recycled from Aqueous phase[a]	g/kg biomass	26.25
P recycled from Aqueous phase[a]	g/kg biomass	10.33
Ammonium Nitrate Input after Recycling[a]	kg/kg biomass	0.08
Triple Superphosphate Input after Recycling[a]	kg/kg biomass	0.02

Data source:
[a] Modeled.
[b] Wu and Liu (2016).
[c] Palou-Rivera and Wang (2010).
[d] Saydah (2015).

2.4.2. Co-products from HTL

When using HTL as the oil conversion technology, co-products including the nutrient-rich aqueous phase, gaseous phase and bio-char, can all be reused within the production system to reduce the primary fertilizer, CO_2 and energy inputs demand by the system (Fortier et al., 2014; Frank et al., 2013). Energy recovery may occur through the combustion of char and bio-crude to generate heat. The nutrient-rich liquid stream can be recycled into the cultivation pond as a nutrient supply for microalgae growth, while the gaseous fraction is composed mostly of CO_2 which can be reused for microalgae cultivation. Detailed modeling assumptions for each co-product are described in the supplementary material.

2.4.3. HTL Co-product treatment methods

Six co-product utilization scenarios and four co-product allocation strategies based on co-products of the HTL process are investigated (Table 3). Recycled nutrients are assumed to displace synthetic fertilizers. Recycled CO_2 gas for microalgae cultivation displaces CO_2 that would otherwise be piped in. The biochar is the only co-product that requires allocation strategies. System expansion methods are the default co-product allocation approach, but economic allocation and energy allocation are also included.

2.4.2.1. Scenario 1: economic allocation. Economic allocation is an alternative approach to displacement calculations; it partitions the impacts of a production system among co-produced products based upon the economic value of each product. In this study, the price of renewable diesel is assumed to have the same market value of conventional diesel of $ 0.78/L (DOE, 2018).

The price of biochar is assumed to be equal to or less than agrichar and charcoal, reported in a large range from $ 0.08 to $ 13.5 per kg. A mean value of $ 2.65/kg biochar was used (Jirka and Tomlinson, 2013; Kulyk, 2012).

2.4.2.2. Scenario 2: energy allocation. Energy allocation is similar to economic allocation, but partitions the impacts based on the energy value of each product. The HHV of biochar and bio-crude oil are used to calculate the energy content in each. In this scenario, the environmental impacts are allocated based on energy content divided between bio-crude oil and biochar, and upgrading of bio-crude oil to renewable diesel is included separately.

HHV of biochar is reported to range from 5 to 15 MJ per kg (Barreiro et al., 2013; Neveux et al., 2014). The HHV of bio-crude oil ranges from 33.6 to 37.3 MJ per kg (Jena et al., 2011; Neveux et al., 2014). A conservative value as 7 MJ/kg is used for HHV of biochar and 35.7 MJ/kg is used for bio-crude oil. The HHV of renewable diesel is assumed to be the same as conventional diesel at 48 MJ/kg. Though the lower heating value (LHV) is a more appropriate indicator of energy content, the LHV for some products were not available, and thus HHV is used.

2.4.2.3. Scenario 3: mass allocation. The mass allocation method partitions environmental impacts based on mass of biochar and biodiesel. The mass of biochar and bio-crude oil resulting from HTL varies under different operation conditions as modeled. The renewable diesel mass is estimated using bio-crude upgrading efficiency at 85% (Saydah, 2015).

Table 3
Scenario description of Co-product treatment for HTL pathway and LE pathway.

Pathway	Products	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5	Scenario 6
HTL	Bio-char	Economic Allocation	Energy Allocation	Mass Allocation	Soil Amendment Displacement	Combusted in CHP[a] to produce Heat and Electricity	Combusted in Boiler to produce Heat
	Aqueous Phase	Recycled	Recycled	Recycled	Recycled	Recycled	Recycled
	CO_2	Reused for Cultivation	Reused for Cultivation	Reused for Cultivation	Reused for Cultivation	Reused for Cultivation	Reused for Cultivation
LE	Glycerol	Economic Allocation Glycerol Price	Displace Glycerol	Displace Glycerol	Displace Glycerol	Displace Glycerol	–
	Algal Cake	Economic Allocation Cattle Feed Price	Displace CA Dairy Cattle Feed (PCDairy Model)	Displace Fishmeal Protein Based	Recycle Nutrients and Energy in AD	Recycle Nutrients and Energy in HTL	–

[a] CHP=Combined heat and power system.

2.4.2.4. Scenarios 4: system expansion. Biochar used as a soil amendment has been investigated for soil fertility improvement and carbon sequestration in agricultural systems and forest systems by numerous studies (Sackett et al., 2014). The composition of biochar varies with the operating conditions and feedstocks, and a typical N content of biochar is 0.5% (Wang et al., 2014). It has been reported that the application of biochar from pyrolysis as soil amendment for corn cultivation increased fertilizer efficiency and crop yields, reduced N_2O emissions from soils by 20%–80%, and increased soil organic carbon (Wang et al., 2014). This study adopts the assumptions from Wang et al. (2014), assuming a onetime application of 30 t/ha biochar on a soil planted with corn would reduce fertilizer application by 10% in the year following and reduce N_2O soil emissions by 30%. Fertilizer inputs for California corn production are used for evaluating the environmental benefits of biochar as soil amendment. The GHG emission from fertilizer application on a typical California corn farm is 270 kg CO_2e/ha with 4.54 kg N_2O/ha (Zhang and Kendall, 2016). Fertilizer input data are adopted from University of California–Davis (UCD) cost and return studies (Brittan et al., 2004, 2008; Frate et al., 2008). The assumptions are simplified without considering biochar quality differences and the fertilizer displacement variability at different locations. The potential for long-term carbon sequestration is not considered.

2.4.2.5. Scenario 5 and 6: recycling and reuse in a closed-loop system. Scenarios 5 and 6 test the effects of using the generated biochar as an energy source within the production system. In scenario 5, biochar is combusted in a combined heat and power (CHP) unit and is assumed to displace natural gas and grid electricity. The efficiency of CHP to convert biochar into electricity and heat is 36% and 50%, respectively. In scenario 6, biochar is combusted in a boiler to produce heat and displace natural gas use only. The boiler operates at 85% efficiency. The energy content in biochar is estimated using the HHV of biochar at 7 MJ/kg (Barreiro et al., 2013; Neveux et al., 2014).

2.5. Microalgae biodiesel production through the LE pathway

Lipid extraction is a widely modeled microalgal biodiesel production pathway. In contrast to lipid extraction from dry biomass, a wet lipid extraction technology is preferred for microalga because it avoids extensive thermal input for drying while still yielding relatively high crude algal oil. The extracted lipid is assumed to be transported and processed in a biorefinery. The algal biomass remaining after LE (algal cake) and glycerol co-produced from transesterification are two co-products that can be used in various applications.

2.5.1. LE pathway modeling

The model of lipid extraction from wet microalgae biomass using hexane extraction is adopted from a previous study (Yuan et al., 2015). Compared to the dry extraction process, wet extraction requires no thermal drying, but does require cell disruption with high pressure homogenization. The electricity and heat requirement for extraction is 0.68 MJ/kg biomass and 2.85 MJ/kg biomass, respectively. The lipid extraction efficiency is 74%. Transesterification is the conversion technology used to convert crude algal oil to biodiesel. With a production of 1 kg dry microalgae biomass, the yields of biodiesel, glycerol and algal cake are 5.75 MJ, 17 g and 0.84 kg, respectively (Yuan et al., 2015). The energy content of biodiesel is assumed 37 MJ/kg (Woertz et al., 2014).

2.5.2. Co-products from LE pathway

Algal cake and glycerol are co-products from the LE and transesterification route. The modeled algal cake is composed of 8% lipid, 39% protein, 43% carbohydrate and 10% ash (dry weight based). This nutrient rich algal cake has great potential to be used for animal feed, fish feed or organic fertilizer; the energy and nutrients can also be recycled and reused in the microalgae cultivation processes through energy recycling technologies. Glycerol is assumed to displace synthetic glycerol with a 1:1 mass ratio, though currently glycerol from biodiesel production is the dominant source in the U.S. market.

2.5.3. LE Co-product treatment methods

As described in Table 3, four utilizations of algal cake are modeled: displacement of dairy cattle feed, displacement of fishmeal, on-site anaerobic digestion (AD) for energy and nutrient recycling, and on-site HTL of biomass residual for energy and nutrient recycling. Glycerol is treated simply in these scenarios; either through economic allocation in Scenario 1, or displacement assuming one to one substitution for synthetic glycerol. The treatment of algal cake is described for each scenario below.

2.5.3.1. Scenario 1: economic allocation. Economic allocation is based on the market price of biodiesel and glycerol, which are $ 0.92/L (DOE, 2018) and $ 0.11/kg (Yuan et al., 2015), respectively. The market price of algal cake is estimated based on the Feed Value Calculator developed by Saskatchewan Ministry of Agriculture (2012) assuming the algal cake is used as cattle feed. The Feed Value Calculator calculates the relative value of crude protein, total digestible nutrients (TDN), phosphorus, calcium and moisture content based on the market price of reference feeds. In the current estimation, the 2017 average price of canola meal and barley grain in US were used as reference. The algal cake was assumed to be sun dried to 40% moisture content before transportation and use. A TDN value for algal cake of 78% was used for price estimation (Mišurcová et al., 2010). The market value of algal cake is estimated as $ 175/t based on its biomass substrate characteristics.

2.5.3.2. Scenario 2: system expansion - displacement of California dairy cow feedstuffs. Based on review of the existing literature, no research or assessment of the displacement value for algal cake in California exists. To conduct this calculation a feed optimization tool tailored to California is identified, PCDAIRY_2015_USA (Least Cost and Ration Analysis Programs for Dairy Cattle), referred to hereafter as PCDAIRY (Robinson and Ahmadi, 2015). PCDAIRY uses an economic optimization based on the price of available feeds to recommend a balanced ration at the lowest cost. To identify feedstuffs likely to be displaced by the introduction of algal cake, PCDAIRY is run with and without algal cake. By doing so, the consequential change induced by introducing algal cake into the feed market in California can be estimated. Of course, if algal cake is introduced in very large volumes, the price of algal cake and competing feeds could change; these displacement calculations implicitly assume that the introduction of algal cake from the simulated facility will not have a significant effect on the price of other feeds. Assumptions and operating parameters that were used in the PCDAIRY tool can be found in the supplementary material.

Table 4 was calculated using PCDAIRY, it reflects a model run with an optimization goal of milk sale profit given fixed nutrient composition and prices for each feed. Based on PCDAIRY calculations, the addition of algal cake in a standard dairy cattle feed ration would result in small changes to all ration constituents but notable increases in corn silage, and decreases in alfalfa hay and dry distiller's grains and soluble (DGS). These changes constitute the effects of adding algal cake to a dairy feed ration and will be used to calculate its displacement value.

2.5.3.3. Scenario 3: system expansion- displacement of fishmeal. Lipid-extracted algal biomass is a suitable candidate to partially replace the use of fishmeal in fish farming. It is found that replacing up to 10% of the crude protein in fishmeal and soybean protein by lipid-extracted algal biomass (including species *Navicula* sp., *Chlorella* sp. and *Nannochloropsis salina*) residual does not lower the growth rate or the feed efficiency in fish farming applications (Patterson and Gatlin, 2013). The displacement ratio of algal biomass to fishmeal in this study is estimated at 0.975 based on protein content (39% for algal cake and 40% for fishmeal). Based on previous LCAs, a primary energy requirement of 19.85 MJ and emissions of 1.35 kg CO_2e are associated with the production of 1 kg of fishmeal (Patterson and Gatlin, 2013; Pelletier et al., 2009).

2.5.3.4. Scenario 4 and 5: recycling and reuse in a closed-loop system. Two recycling technologies, AD and HTL, are tested for scenarios 4 and 5. AD produces biogas, suitable for use in a CHP unit, and digestate, from which the liquid fraction is recovered and fed into the ORPs for water and nutrient recycling, and the solid fraction is composted and used off-site as a nutrient-rich soil amendment.

Just as when HTL is used to process whole microalgae, HTL applied to algal cake produces a CO_2-rich gaseous stream, a nutrient-rich aqueous stream, a biochar and a bio-crude oil product. The nutrient rich stream is used for nutrient recycling while bio-crude oil and biochar are combusted in a boiler for heat generation. The results for Scenario 4 and 5 are adopted from previous study by Zhang et al. (2014).

Table 4
California Dairy Feed Rations with Algal Cake Addition ($175/t, dry matter based).

	No Algal Cake	With Algal Cake
Algal cake (kg/d)	0.00	1.36
Corn silage (kg/d)	3.89	4.44
Wet GDS (kg/d)	3.79	3.81
Barley (kg/d)	5.50	5.88
Alfalfa hay (kg/d)	4.68	4.14
Almond hulls& shell (kg/d)	3.03	3.04
Dry DGS (kg/d)	2.72	0.23
Beet pulp (kg/d)	0.00	0.80
Dicalcium phosphate (kg/d)	0.07	0.00
Limestone (kg/d)	0.05	0.11

2.6. Data sources

The primary data for modeling parameters such as the microalgae growth model, energy inputs for cultivation, harvesting and HTL and upgrading inputs, are based on peer-reviewed literature as described in each section. The reference LCI data including fertilizer production, glycerol production, hydrogen, grid electricity and natural gas production and related emissions used in the model come from the Gabi Professional database (Thinkstep, 2016) and the Ecoinvent database (Ecoinvent Center, 2016). The US Western grid electricity mix is used for electricity inputs. LCI data are provided in supplementary material.

3. Results

3.1. HTL pathway performance without Co-product allocation

The effects of operation conditions on renewable diesel yield, primary energy consumption and GWP100 of the system before

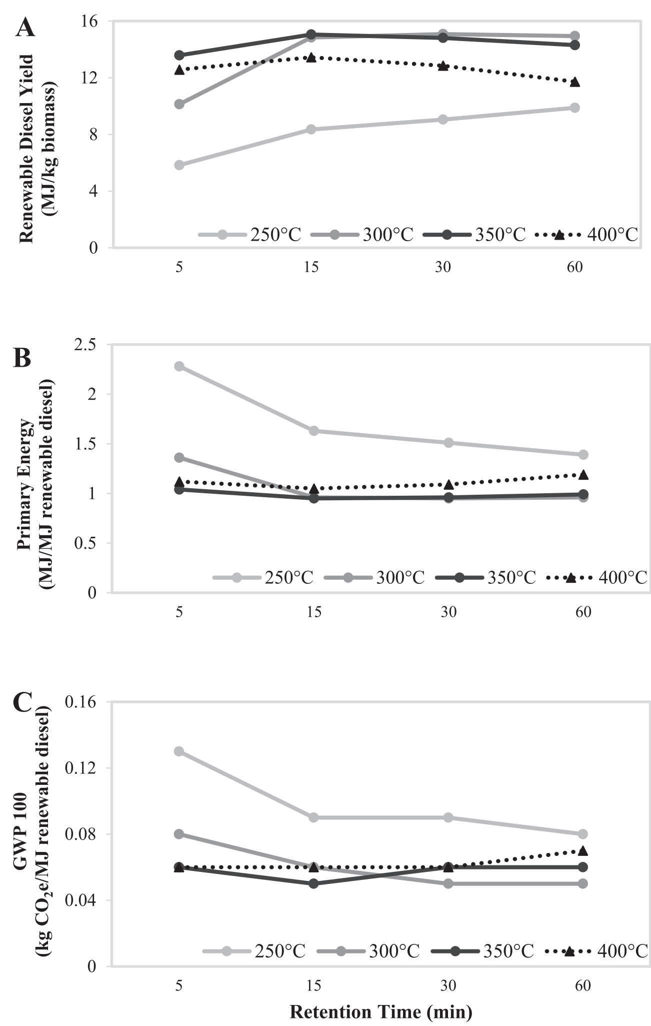

Fig. 2. Effects of operation conditions on renewable diesel yield, GWP100 and primary energy consumption.

Table 5
Life Cycle GHGs and Energy by Process per MJ Renewable Diesel Production without co-product allocation. HTL was modeled at 350 °C for 15 min.

	Cultivation	Harvesting &Dewatering	HTL	Upgrading	Recycled Nutrients	Sum
Primary Energy (MJ/MJ)	0.76	0.24	0.25	0.11	−0.17	1.18
Fossil Energy (MJ/MJ)	0.59	0.20	0.25	0.11	−0.15	1.00
GWP_{100} (g CO_2e/MJ)	52.53	10.70	10.86	2.29	−14.11	62.27
GWP_{20} (g CO_2e/MJ)	57.14	12.27	13.25	2.80	−14.99	70.47

allocation are shown in Fig. 2. Among all tested conditions, the yield of renewable diesel is the highest at temperatures of 350 °C for 15 min. The lowest primary energy consumption and life cycle GHG emissions from 1 MJ renewable diesel production occurred at temperatures of 300 °C and 350 °C with retention time from 15 min to 60 min. Operating at 350 °C for 15 min is used as the optimal condition because a shorter retention time is preferred for lower cost at industrial facilities. The following sections report results using this operation condition as default.

Table 5 shows process-based contributions to energy and GWPs. Cultivation is the most energy intensive stage for renewable diesel production, due to the electricity use for CO_2 pumping, mixing, and fertilizer inputs. The harvesting and dewatering processes, HTL process and upgrading stage contribute 18%, 18% and 8% of total primary energy consumption of the system, respectively. Nutrients recycling from the aqueous phase reduces the system primary energy by 13%. Before allocation of co-products, the total primary energy input for renewable diesel is 1.18 MJ/MJ, and the GWP100 for renewable diesel is 0.062 kg CO_{2e}/MJ. The H_2 input is a key variable for bio-crude oil upgrading that affects the primary energy consumption and GWP of the renewable diesel. The demand of H_2 is determined by the bio-crude oil properties and it affects final fuel yield and quality (Wang et al., 2017). This study makes a simple assumption with the H_2 requirement of 3%wt of bio-crude oil, however, the uncertainty of H_2 demand variation driven by HTL operation conditions and bio-crude oil properties requires careful estimation.

3.2. Effects of Co-product treatment on the HTL pathway and LE pathway

Fig. 3 reports the results for un-allocated energy and emissions from the HTL pathway and LE pathway along with results from different co-products treatment scenarios.

For the case of HTL pathway, economic allocation leads to the lowest energy and life cycle GHG intensity (or carbon intensity) for renewable diesel among all allocation approaches because of the high value estimated for biochar. When the price of biochar is set at $ 0.5/kg instead of $ 2.65/kg (default value), the economic allocation results in approximately equal carbon intensity of biochar to other allocation methods. Second to economic allocation in terms of favorable carbon intensity is the substitution of biochar for soil amendments. Depending on the long term carbon sequestration potential of biochar in soils, this use could result in even lower carbon intensity. In terms of closed-loop utilization, combustion in a CHP is slightly preferable to combustion in a boiler for heat generation only. Overall, the allocation approach has relatively small effects on the final results due to the small yield of biochar from HTL. This suggests the findings for renewable diesel produced through the HTL pathway are reasonably robust to changes in the value of co-products and the allocation method chosen.

Without allocation of co-products, biodiesel production from LE requires much higher energy (3.52 MJ/MJ) than renewable diesel from HTL, because the yield of crude algal oil from 1 kg biomass under the LE pathway is less than the crude algal oil produced under HTL. However, biodiesel is very sensitive to the treatment of algal cake and allocation strategies due to the large quantity of algal cake production (detailed results can be found in the supplementary material). For biodiesel production, using algal cake as feed (scenarios 1, 2 and 3) show higher environmental benefits than closed-loop nutrient and energy recycling scenarios (scenario 4 and 5). There are large uncertainties related to the algal cake treatment, such as the price, the nutrient content, the feasibility to use as animal feed, and perhaps additional processing.

Comparing the recycling strategies of co-products in a closed-loop and selling co-product in an open-loop system, a closed-loop system design avoids the allocation process and results in fewer uncertainties of environmental impacts, while the drawback is the loss of potential economic value (as well as the environmental best-use) from co-products. In general, the HTL pathway results in more consistent environmental performance results and is subject to fewer effects from co-product treatment strategies. This is because HTL yields a very small quantity of co-product (biochar) that can be used outside the production system, reusing most non-fuel products within the system. While the LE pathway exhibits higher uncertainty, it may also hold promise for higher profits from selling the high value algal cake as animal feed, as illustrated in Fig. 3 under the bars for Economic Allocation.

4. Uncertainties and discussion

4.1. Uncertainty of Nutrient Recycling Capacity on HTL pathway

Microalgae cultivation with recycling of the aqueous phase and gases from HTL may introduce heavy metals and inorganic contaminants into the growth media. However, there are no consistent estimates of nutrient content in the aqueous phase, nor are there studies that have definitively proven the feasibility of recycling the aqueous product to the ORP without affecting microalgae growth performance due to different experimental conditions and limited data (Liu et al., 2013; López Barreiro et al., 2014). To better estimate the effects of nutrient recycling rates used in the ORP, three recycling rates for N and P from the HTL aqueous phase are tested: the low rate assumes 15% of total input N and 20% of total P can be reused for cultivation; the default rate assumes 50% of total N and 80% of total P can be reused; and the high recycling rate assumes 95% of total N and 95% of total P can be reused for cultivation. Effects on the HTL production system (before co-product treatments) are shown in Fig. 4.

Without allocation of co-products, HTL system GHG emissions range from 72 g CO_{2e} to 52 g CO_{2e} to produce 1 MJ renewable diesel from the low rate case to high rate case; while the total energy input ranges from 1.31 MJ/MJ to 1.09 MJ/MJ. The high recycling rate could result in a substantial reduction of primary energy inputs and GHG emissions by 20% and 30%, respectively. Comparing to the co-product bio-char from HTL pathway, the recyclable nutrients could contribute to higher environmental benefits. Therefore, the impact of heavy metals and inorganic contaminants on microalgae growth and the fate of heavy metals need to be better understood in order to evaluate the potential or limits on recycling HTL products.

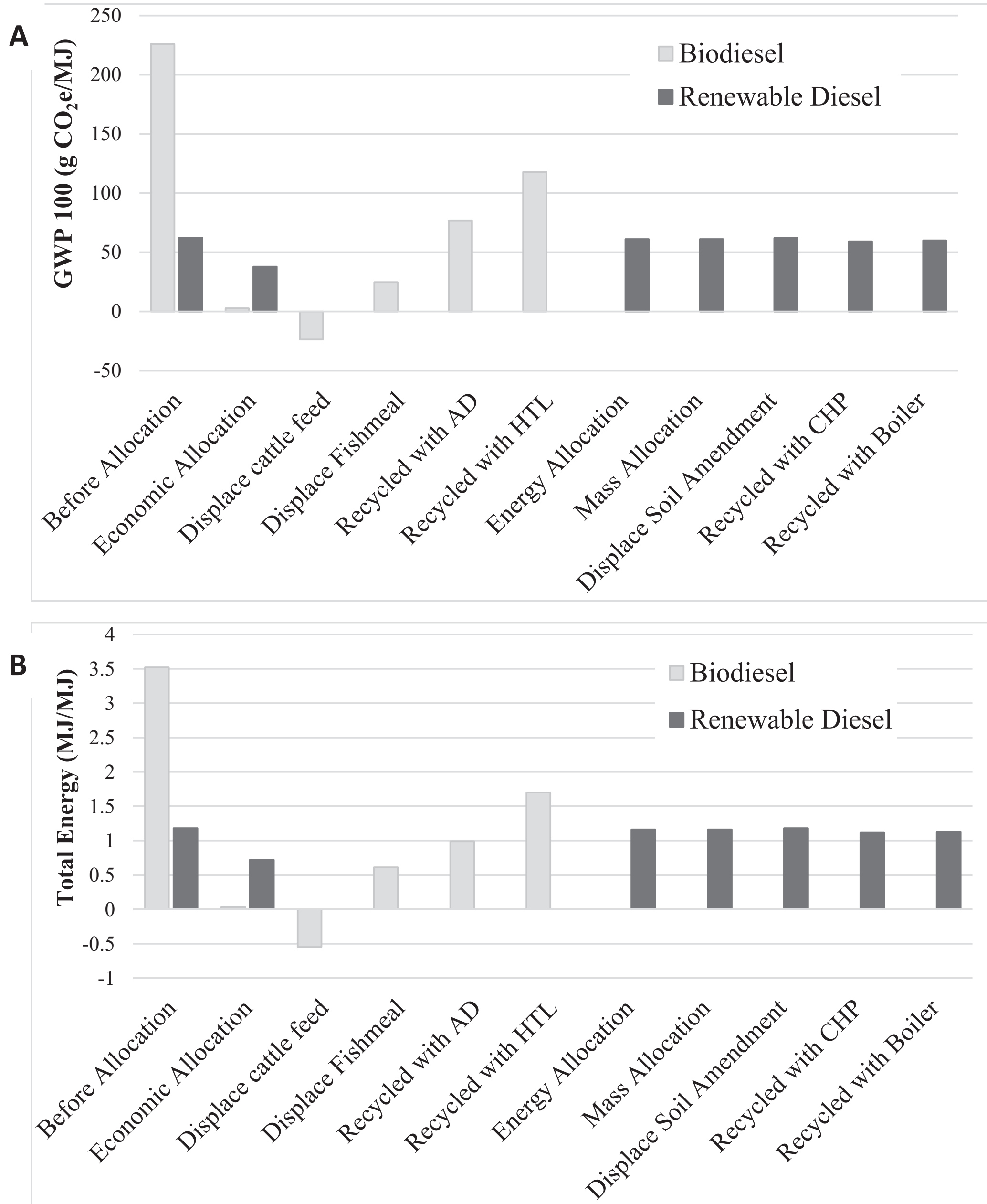

Fig. 3. GHG emissions (A) and Total Primary Energy (B) for Biodiesel and Renewable Diesel Production with Co-product Treatment. For reference, GHGs from petroleum diesel is approximately 95 g CO_2e/MJ.

4.2. Uncertainty of biochar price and displacement effects on HTL pathway

Although biochar has been recommended as a soil amendment for improving soil properties, the potential for biochar application to reduce fertilizer demand for crops produced on amended soils is uncertain due to variations in biochar properties, as well as different field and climate conditions (Glaser et al., 2015). Similarly, the price of biochar is uncertain, as discussed previously. Both of these uncertainties have implication for co-product allocation calculations that determine the impact attributable to renewable diesel produced through the HTL pathway. To understand the potential effect of these uncertainties on the results for renewable biodiesel, a sensitivity analysis is conducted on the fertilizer displacement ratios attributed to biochar and its market price. Three scenarios are tested: a low quality scenario that assumes the biochar application reduces fertilizer inputs by 10% and reduces N_2O emission by 5% with a price of $ 0.2/kg; a default scenario that assumes biochar application reduces fertilizer inputs by 20%, N_2O emission by 10%, at a price of $ 2.65/kg; and a high quality biochar scenario that assumes a reduction of 80% of fertilizer demand and 20% of N_2O emissions, at a market price of $ 13.5/kg.

Results show that the price of biochar significantly effects the GHG emissions and energy inputs of renewable biodiesel (see Fig. 5). The GHG emissions range from 14.7 to 54.9 g CO_2, while the total energy of per MJ renewable diesel ranges from 0.25 to 0.94 MJ. However, when varying biochar qualities are evaluated using displacement calculations, such effects are not seen. The high quality biochar with high fertilizer reduction rate does not significantly improve GHG emissions and total energy of renewable

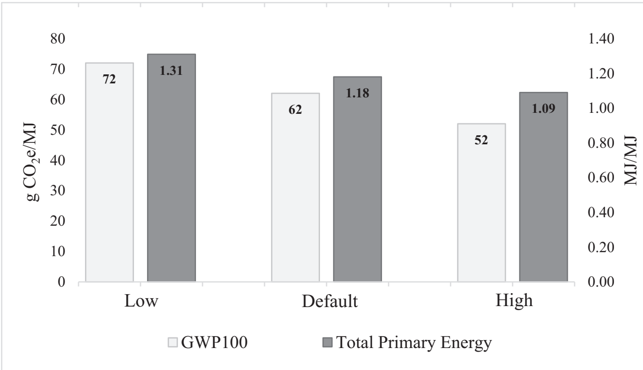

Fig. 4. Effects of Nutrient Recycling Capacity on GHGs and Energy per MJ Renewable Diesel Production (Before co-product treatments).

diesel. The impacts from different fertilizer displacement ratios are relatively weak, which is the result of the high application requirement of 30 t/ha of biochar in the field. It should be noted that this displacement calculation only considers biochar displacing fertilizer, and excludes other potential benefits from biochar including increased crop yield, improved soil properties, soil carbon sequestration effects and substitution of mineral fertilizer nutrients.

4.3. Uncertainty of algal cake price on LE pathway

Sensitivity analysis of life cycle displacement credits of algal cake at different prices is conducted to understand the potential effect. At lower prices, algal cake offsets more GHG emissions and energy inputs, meaning the credit attributed to the algal biodiesel production system is higher (Fig. 6). At a lower price, algal cake displaces larger quantities of dry DGS in the feed ration, which has a higher market price and involves higher environmental impacts to produce (as shown in supplementary material). This sensitive response of environmental impacts to prices is critical to the life cycle performance of biodiesel produced from LE pathway. However, estimating the market price of algal cake as feed is challenging to this research, because algal cake is not yet a commercial product in the feed market. Moreover, algal cake may concentrate chemical elements which can be toxic to animal and human health, depending on microalgae species, cultivation or conversion processes. Thus, the feasibility of using algal cake used for feed still requires further research.

5. Conclusion

This research explores the real, method-induced, and model-induced variability of co-product handling strategies for microalgal biofuels by comparing two production pathways: renewable diesel from HTL and biodiesel from LE. Before co-product allocation,

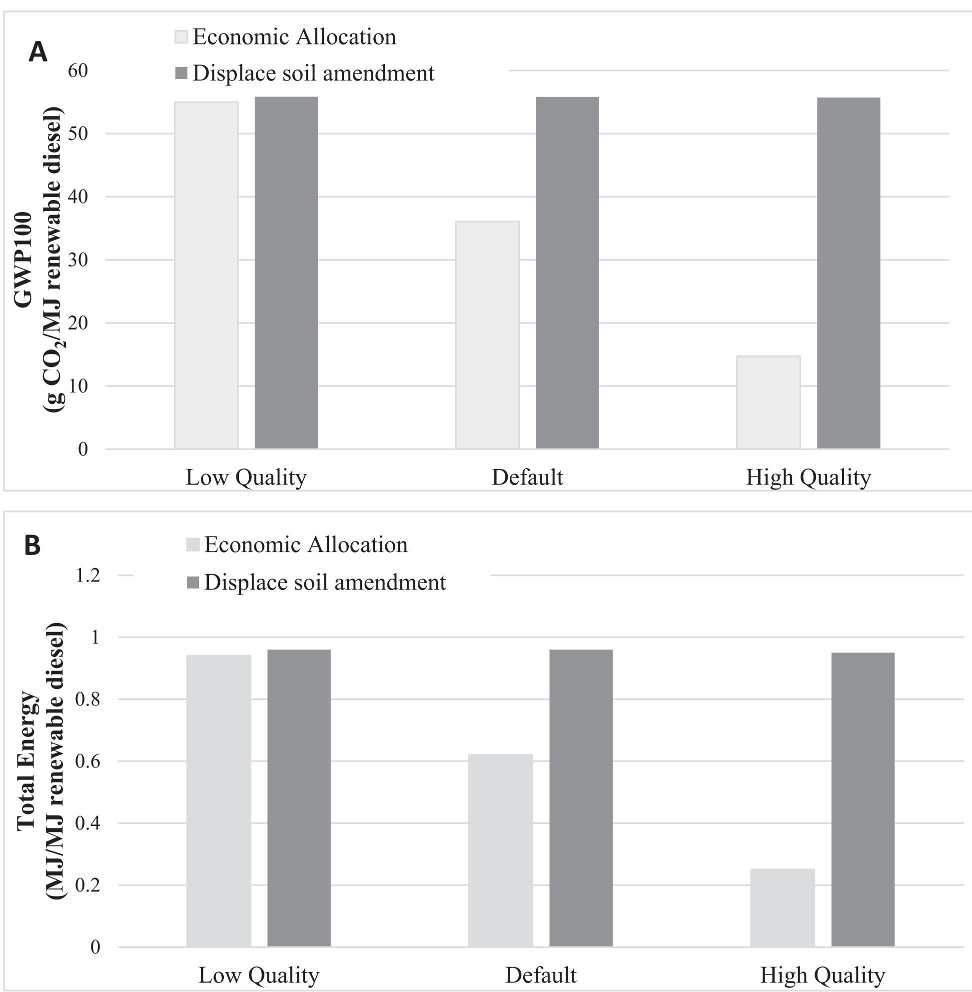

Fig. 5. Sensitivity Analysis of CO_2e Emissions and Total Energy for Renewable Diesel affected by Biochar at Different Prices and Displacement Effects.

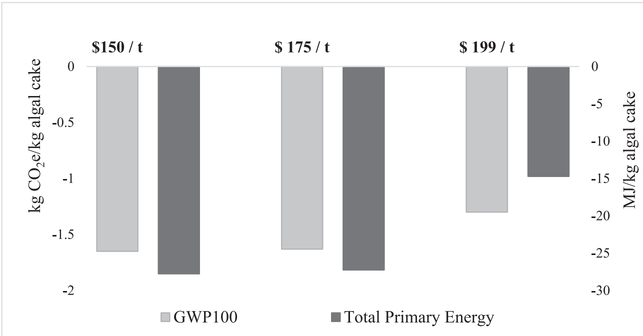

Fig. 6. Sensitivity Analysis of Avoided CO_2e Emissions and Total Energy by 1 kg Algal Cake at Different Prices.

the GHG emissions from renewable diesel (HTL) and biodiesel (LE) were 62 g CO_2e/MJ and 226 g CO_2e/MJ, respectively. After allocation, the carbon intensity of renewable diesel varied from 38 g CO_2e/MJ to 62 g CO_2e/MJ, while the carbon intensity of biodiesel had a dramatic range from −24 g CO_2e/MJ to 118 g CO_2e/MJ. Not surprisingly, a comparison of these two pathways subject to a variety of scenarios that varied the co-product utilization strategies and allocation methods, suggest that more robust carbon intensity estimates are achievable when co-products have little contribution to the performance of the biofuel, or when they are internally recycled.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.05.137.

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