



Environmental profile of algal Hydrothermal Liquefaction – A country specific case study



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ABSTRACT

Microalgae are known to be an important feedstock not just for biofuel but also for biochemical production. In this investigation we utilise a cradle-to-biorefinery-gate attributional LCA (aLCA) methodology to evaluate the environmental impacts of *Nannochloropsis* sp. derived algal biocrude production. A database of primary experimental data for continuous fast Hydrothermal Liquefaction (HTL) and Hydrotreating (HDT) is combined with secondary data from literature to investigate the overall environmental profiles of cultivation, dewatering, HTL and HDT for various scenarios based on the energy generation mix of 5 countries (Brazil, UK, Spain, China and Australia) as well as a comparison with fossil crude. The investigation found that Brazil delivers best environmental profiles for all scenarios, primarily due to its significant contribution from hydropower. Furthermore, the cultivation and HTL processes account for nearly 90% of environmental burdens whereas dewatering and HDT only contribute less than 8%. The research findings highlight the importance of the several factors on the resulting 3G biofuel profiling e.g. energy resource, processing technology choice and the co-product(s) and emissions profiling methodology. Algal biocrude is still undergoing research and development compared to the well-developed fossil crude industry. Via integration and optimisation at process and value chain levels, algae-derived biocrude has the potential to deliver an environmentally sustainable alternative to the fossil crude, provided that the energy input for processing is from a renewable source.

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1. Introduction

The use of fossil fuels to sustain global energy demand has resulted in detrimental environmental effects. As a major energy consumer, transportation sector alone accounts for 32% of total EU energy demand and in 2012 was responsible for 25% of greenhouse gas (GHG) emissions in the EU-28 [13,14]. The increasing GHG levels (elevation in CO₂ concentration to 400 ppm) along with fossil resources depletion have triggered ambitious policies mandating renewable energy sources within regional/national energy portfolio [17]. The role of biofuels in the UK is acknowledged under the EU Renewable Energy Directive (RED), mandating a policy target of a 10% share of renewable energy within the EU transport framework by 2030 [15]. Currently, 1st and 2nd generation biomass feedstocks are commercially used for fuel production but due to their intrinsic environmental and social disadvantages [12], emphasis on 3rd generation (3G) feedstock such as microalgae has resulted in substantial research and development.

Compared to other biomass feedstock, microalgae have a faster growth rate, ability to grow in fresh, saline or waste water and more importantly, it is not a primary food crop and thus does not compete for arable land or increase food prices [41]. Although algae compete with food

for fertiliser, the nutrient requirements can be potentially offset by algal cultivation using waste water as well as nutrient recycling from aqueous phase. The lipid, carbohydrate and protein present in microalgae can be converted to fuel precursors or chemicals using several processing technologies [43].

For conversion of algal biomass without prior fractionation, Hydrothermal Liquefaction (HTL) is understood to be a promising treatment option, particularly since it negates drying thus reducing costs and energy consumption [31]. Water under hydrothermal conditions undergoes an enhancement in its properties (density, C_p, ionic product and dielectric constant) causing it to behave like an organic solvent with acidic/basic catalytic properties which is ideal for the conversion of biomass macromolecules to simpler components without addition of harmful solvents [49]. Several studies [5,21,26,47] have successfully demonstrated utilisation of HTL to convert algae paste and the research outcome has pointed towards reducing the Residence Time (RT) from hours to minutes [18,25,40]. Typically, HTL alone is not sufficient and post processing of HTL derived biocrude is necessary to reduce the Oxygen/Nitrogen content through Hydrotreatment (HDT).

The implication of combining these processes on environmental performance is expected to be significant especially since there have been major improvements in cultivation and dewatering. It is necessary to examine whether advances in algal biofuel production process are cascaded to result in improvement for its environmental profiling. A widely

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accepted method used to quantify the environmental metrics of a product/process is Life Cycle Assessment (LCA). There have been several investigations which attempt to quantify LCA impacts of algal biofuels production via different processing routes with most focusing on cultivation or biodiesel production using a lipid extraction pathway [39, 43]. The outcome from these studies elucidated that algae drying process is a main contributor to GHG emission and thus, there has been increasing research attention on a wet processing route where algal biomass with high moisture content can be directly utilised as feedstock thus there is no need to dry feedstocks [9].

To date, a limited number of LCAs have been conducted on HTL (or thermochemical) processing [4,11,19,20,22,32,44]. Most studies are based on literature data and therefore the general consensus on LCA profiling has not yet been attained. For instance, Frank et al. [20] concluded that even though HTL uses wet algae, there is an increase of GHGs emission compared to the traditional route (drying and extraction). But conversely [19] suggested that with proper heat integration it is possible to reduce GHGs by 76%. Additionally, by using reaction conditions which maximise biocrude yield (low RT and high temperature) coupled with the high energy recovery in the biocrude of up to 88% (Higher Heating Value (HHV) on a dry weight basis), a further reduction in GHGs and costs can be obtained, resulting in environmental and financial benefits [56]. Literature review indicates that no publically available study has used data from a continuous HTL flow reactor system which would most probably be implemented at commercial scale.

Ultimately, scale-up of an algal biorefinery with multiple product vectors would be the way forward to support (economically) algal biofuel production, but nonetheless, the biofuel produced should instil environmental benefit. Depending on the geographic location or strain used, various products could be extracted and produced using benchmarked processing techniques, for instance HTL biocrude production [57,58]. The algal biorefinery system with energy inputs met by national/regional grid energy mix might not lead to environmentally superior algae biofuel compared with fossil fuels which depends on the region under investigation. A thorough literature review suggests a knowledge gap in LCA of continuous HTL processing (and subsequent HDT) of algal biomass with varying system configuration (e.g. low RT) and different locations [39,43]. Particularly there is a lack of country specific comparison analysis of HTL-based algal processing based on empirical data derived from experimental work. This study presents LCA modelling of potential algal biofuel refinery systems to advance the understanding of environmental profiles of algal HTL and HDT processing with varying HTL/HDT reaction configurations and (energy) supply chain at different locations. This study aims to not only fill up the knowledge gap, but more importantly provide scientific insights into the process design and optimisation to enable ongoing empirical work to be more efficiently focused on key environmentally damaging steps to contribute to future sustainable development of algal biorefinery. As such the biorefinery systems are modelled in 5 countries (Brazil, UK, Spain, China and Australia) with differing energy mixture and varying HTL/HDT reaction conditions.

2. Method

2.1. Product system and functional unit

A cradle-to-biorefinery-gate attributional LCA (aLCA) approach was applied to evaluate the environmental impacts of the algae-derived liquid fuels (3G biofuel). As illustrated in Fig. 1, the subsystems modelled within the system boundary include algae cultivation, algae dewatering, HTL and HDT. The aqueous phase fraction produced at the HTL stage is sent to a waste water treatment anaerobic digestion (AD) unit. The biogases produced from AD were further sent to combine heat and power (CHP) system for energy recovery. LCA inventory was developed using primary data obtained from laboratory experiments (for downstream processing HTL and HDT stages), supplemented by secondary

data from literature. However, lab-scale data may not represent the system performance at pilot-scale or commercial-scale fully, given the substantial number of parameters still undetermined. Thus further research would be needed on system scaling-up by process simulation, data production and validation against larger-scale operational data, if available. The LCA study was performed using SimaPro® 7.3 (PhD version) and the biofuel produced from algae, called biocrude, was modelled as potential replacement or blending fuel with crude oil at a refinery and was thus compared directly with fossil crude. The functional unit was defined as 'per MJ crude oil produced at refinery gate'. A problem oriented (midpoint) approach – CML 2 baseline 2000 (v2.05) was applied in the current study as the 'default' Life Cycle Impact Assessment (LCIA) method. The impact categories to be investigated include biotic depletion, global warming potential, acidification, eutrophication, ozone depletion, photochemical oxidation and human and eco-toxicities. Other environmental impact categories including land occupation/land use, Energy Return on Energy Investment (EROEI), are excluded from the current LCA system boundary but could be explored in future research. Especially EROEI, as an indicator to assess the energetic profitability of a system, has been applied in algal biocrude research to explore issues like maximising the energy recoveries of alternatives processes e.g. study carried out by Tercero et al. [59]. EROEI could be incorporated into future LCAs as an energy payback efficiency indicator for algal biocrude production via HTL routes.

The acidification characterisation model incorporated in CML 2 baseline 2000 is derived from RAINS model (Regional Air Pollution Information and Simulation) where main acidifying gases accounted for include SO_x, NO_x, NH₃. However, another concern is the ocean acidification effects caused by CO₂, which is still in its infancy. Thus further studies are required to explore biological and biogeochemical consequences of an ocean acidification process as well as potential incorporation of ocean acidification evaluation into an LCA framework.

2.2. Allocation approach

A 'system expansion' allocation approach was applied for the biocrude production processes to account for the multiple product mixture present in the system. These were 1) HTL stage where multiple-products include the biocrude oil plus electrical power generated from the on-site AD/CHP system in addition to the nutrient contained in recovered biochar; 2) HDT stage where the upgraded biocrude and biochar recovery as a potential fuel source are produced. It was assumed that the electricity co-product would directly displace an equivalent amount of electrical power generated from the average national grid mixture of the corresponding country in each scenario. The biochar recovery from HTL and HDT was assumed to substitute a 'functional equivalent' quantity (dry basis) of national average N fertiliser production and an equivalent amount of charcoal, respectively. Thus, this allocation approach awards the biocrude production system with 'avoided burdens' credits for the fossil fuel consumption and subsequent emissions avoided for an equivalent amount of avoided product generation.

An alternative allocation approach – energy allocation – recommended by the EU RED was examined in sensitivity analysis (Section 3.5) where the environmental burdens were allocated among the co-products (e.g. biocrude and energy recovery) based on their energy contents.

A stoichiometric carbon-counting approach was used to 'track' the biogenic carbon flows from algae biomass into biocrude oil over the life cycle [23]. This C-counting approach with regard to the biocrude was applied to firstly determine the carbon 'sequestered' into the biocrude (from the algae cultivation phase of the life cycle) and downstream release of this carbon during the subsequent processing of the biocrude. The sequestration of carbon into biocrude thus represents a

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