

## The effect of functional unit and co-product handling methods on life cycle assessment of an algal biorefinery

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## **Abstract**

Microalgae are a promising feedstock for sustainable fuel and nutritional products. To be economically viable, algal biorefineries will need to produce multiple products, such as nutraceuticals and animal feed, in addition to fuel. Life cycle assessment (LCA) has been applied widely to algal biorefineries, but results vary across studies. Here we address two methodological challenges that can lead to variability in LCAs of multi-product algal biorefineries: the choice of functional unit and the method used to handle co-products. LCA was conducted for an algal biorefinery that produces two representative products— fuel and animal feed—using three functional units: (1) 1 MJ fuel, (2) 1 kg animal feed, and (3) 1 ha of production area coupled with the system expansion method. For the fuel-based functional unit, the system expansion method was compared to impact allocation based on mass, energy, and market value of the biorefinery products. The choice of functional unit affects the results for several environmental indicators. For ecosystem quality, the choice of functional unit can change the overall balance of environmental impacts from harmful to beneficial. The effect of functional unit on climate change depends on the boundary considered in the model—i.e., “well to gate” or “well to wheels”. When a fuel based functional unit is used, the choice of co-product handling method (i.e., allocation versus the system expansion method) affect environmental impacts, such as ecosystem quality and climate change, differently. LCAs that use a fuel based functional unit to compare algal biofuel to other fuels, multiple co-product handling methods should be analyzed and presented to stakeholders. A land based functional unit coupled with the system expansion method can be used to improve LCAs that compare among multi-product algal biorefineries.

## **Keywords**

Algae, biorefinery, Life cycle assessment, functional unit, co-products

## **1. Introduction:**

Microalgae (herein referred to as algae) are a promising feedstock for fuel, food, nutraceuticals, and other products. Numerous studies have been published on life cycle assessment (LCA) of algal biofuels and animal feed in an effort to quantify their environmental impacts [1–5]. The results of these studies are highly variable, due to the variety of algal cultivation systems, conversion pathways, product distributions, and methodological challenges associated with life cycle modeling. One methodological challenge that leads to variability in LCA results for multi-product systems, such as algal biorefineries, is the choice of a functional unit and its effect on allocation of environmental impacts among products.

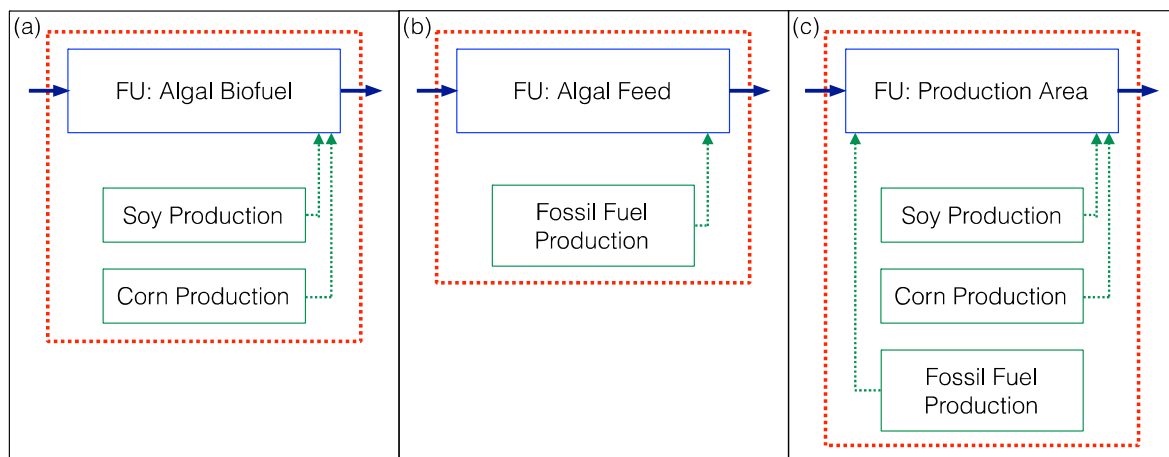
The functional unit plays an important role in LCA and can affect comparative performance of modeled systems. A functional unit represents a quantitative description of the functions provided by the system under study [6]. Its purpose is to provide a reference to which all environmental flows are normalized, which allows a comparison of LCA results on one basis [7,8]. If an LCA modeler does not accurately identify the function of a modeled system, an incorrect choice of FU may follow. A misleading choice of functional unit can, in turn, lead to incorrect results and conclusions about the environmental performance of a process under study. A correctly defined functional unit is more likely to result in an LCA with an “apples-to-apples” comparison of the modeled scenarios. LCA standards, as described in the ILCD handbook [9], as well as ISO 14040 and 14044, specify that the functional unit should be consistent with the goal of the study and all functions of the system [7,8].

Methods that incorporate LCA methodology (e.g., product environmental footprint (PEF), organization environmental footprint (OEF), and the greenhouse gas (GHG) protocol) provide guidance on choosing functional units, and some organizations (e.g., renewable energy directive (RED)) specify the functional unit (e.g., 1 MJ of fuel) [10]. None of these specifications or recommendations provide adequate guidance for modeling biorefineries (e.g., algal systems) with multiple products [10]. Furthermore, for LCAs of multi-product systems, the correct choice of functional unit is not straightforward. Because biorefineries most often produce multiple products, LCA practitioners have taken alternative approaches to address the question of the most appropriate functional unit. Functional units used in LCAs of terrestrial and algal biorefineries have been associated primarily with outputs (e.g., 1 MJ of liquid fuel) [1–3], but also with inputs, e.g., energy content of feedstock [11] and area of agricultural land [12]. Ahlgren et al. recommend using ‘1 biorefinery unit’ as the functional unit for LCA models of terrestrial biorefineries with multiple functions [10]. Most LCAs of algal systems focus on biofuel production and use a functional unit of 1 MJ of fuel. Specifically, 49 out of 55 algal LCA studies analyzed by Tu et al. used fuel or distance traveled based functional units [3]. The benefit of this method is that it enables a direct comparison with petroleum fuels on a per-MJ basis. But to be commercially viable, biorefineries will have to produce high value products (e.g., aqua feed, and nutraceuticals, such as omega-3 fatty acids), in addition to biofuels [5,13–15]. In such cases, the function of the system is not to only produce fuel, but also one or more valuable co-products, and, therefore, the functional unit should be modified to account for all products produced by the biorefinery. The use of different functional units, i.e., for each alternative high-value product studied, however, will limit the ability of practitioners to compare life cycle environmental impacts among studies of algal systems.

Allocating environmental impacts among services provided by multi-product systems is one of the most studied methodological issues in LCA [6], and for biorefineries it is tied closely to the choice of functional unit [16]. Environmental impacts of multi-product biorefineries can be dealt with in two ways. The first method, allocation, divides environmental impacts among products based on their mass, energy, or economic values [7,8]. But allocation may skew LCA results for multi-product biorefineries. If the primary economic driver for a system is a product that is a small fraction of the total mass produced by the system (e.g., astaxanthin from algae), allocating impacts by mass or energy will assign only a small impact to the primary product. Furthermore, for biorefineries, allocation based on energy results in different conclusions compared to allocation based on mass or market value [17–19]. Volatile fuel prices and government subsidies can introduce challenges in choosing correct values for allocation based on market values [20]. These challenges suggest that allocation may not be the best option for modeling multi-product biorefineries. The second method, the “system boundary expansion method” (also called the “displacement method” or the “substitution method”), is applied by expanding the model boundary to include replacement of conventional products by the biorefinery’s co-products. However, coupling product-based functional units with the system expansion method presents challenges when it is unclear which biorefinery product is the main product and which is the co-product [18]. For an algal biorefinery that produces fuel and feed, both products represent major components of the algal biomass in terms of mass, energy, and economic value.

ISO 14044 states that whenever possible, the system expansion method should be used, and the allocation of life cycle environmental impacts among products should be avoided [9,21]. But

ISO also states that when multiple allocation schemes may be applicable, LCA should be performed with all allocation schemes and their results compared. The system expansion method can be applied to an algal biorefinery that produces biofuel and animal feed (as two example products), in two ways. First (in the most common approach) biofuel is designated as the main product and functional unit, and the modeled system is credited with avoided impacts resulting from the displacement of a conventional product (e.g., corn- and soy-based animal feed) that the co-product (defatted algal biomass) replaces (Figure 1a). Second, animal feed is designated as the main product and functional unit, and the modeled system is credited with avoided impacts resulting from the displacement of a conventional fuel product (e.g., fossil oil) that the biofuel co-product replaces (Figure 1b). Although government regulatory agencies have relied primarily on the system expansion method for assessing environmental impacts of terrestrial biofuels [18], there is still no consensus about the correct way to attribute environmental impacts for biorefineries with multiple products [18–20,22,23]. Using a land-based functional unit of 1 ha designates both products, oil and defatted algae, as co-products, and the modeled system is credited with avoided impacts resulting from the displacement of conventional fossil fuel and conventional animal feed (Figure 1c) [5,24]. Land is often used as a reference measurement for agricultural systems with crop yields reported on a per hectare (ha) basis. Since algae can be considered a competitor to conventional agricultural crops (for bioenergy or food purposes), we propose to evaluate its environmental impacts on a per unit of area basis.



Laurens et al. propose that a uniform approach should be taken for LCA and technoeconomic analysis models of algal products, and that a standard set of environmental indicators should be chosen to assess the sustainability of algal biorefineries [25]. The overarching goal of this study is to illustrate challenges associated with the choices of functional unit and co-product handling methods for LCA of multi-product algal biorefineries. We modeled a biorefinery with two representative products—fuel and feed—to illustrate an algal biorefinery with multiple products (e.g., fuel, feed, nutraceuticals, pharmaceuticals). We compare the LCA results for three functional units, two of which are associated with the biorefinery’s products: (1) 1 MJ of biofuel produced, (2) 1 kg of animal feed produced, and (3) 1 ha of cultivation area, using the system expansion method. For the fuel-based functional unit we compared the system expansion method to impact allocation based on mass, energy, and market value of the biorefinery products.

## **2. Materials and Methods**

### *2.1. Modeling Framework*

The algal biorefinery modeled in this study includes algae cultivation, harvesting, dewatering, and processing to bio-crude oil (fuel), and animal feed. The process systems modeling framework developed previously [5,24] was adapted to conduct an LCA of a biorefinery with three functional units: (1) 1 MJ of fuel, (2) 1 kg of animal feed, and (3) 1 ha of production area. The first two functional units were chosen as they represent major outputs and functions of an algal biorefinery. Animal feed was long thought to be a secondary function of algal biofuel production, but given its importance in terms of mass, energy, and economic value, it is as important as fuel [5,26]. The production-area-based functional unit was chosen because it represents land being used to produce all usable products in an algal biorefinery. The system expansion method was used to account for coproducts for these three functional units.

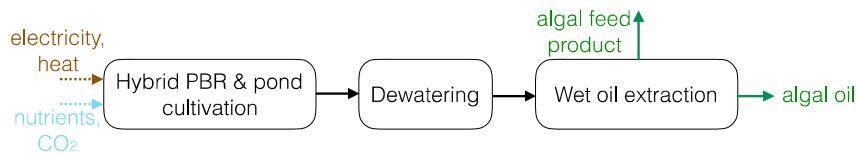
For an LCA model with a functional unit of 1 MJ of fuel, allocation of environmental impacts based on mass, energy content, and market value of oil and post-extraction algae meal as animal feed was compared to using the system expansion method to handle co-products. For mass, the impacts were allocated between oil and animal feed, based on the mass fractions of products, which were 0.27 and 0.73 for oil and defatted algae, respectively. For energy, the impacts were allocated between oil and animal feed based on the energy contents of each product produced by the refinery. We assumed a lower heating value (LHV) of 44 MJ/kg for the oil, and 12.5 MJ/kg for the defatted algae (to be used as animal feed) [27]. Combining the LHV values of each product with their associated masses resulted in allocation factors of 0.57 and 0.43 of the energy produced as oil and animal feed, respectively. For market value, we used the average selling

prices of soy oil (\$87 per kg) and soy meal (\$40 per kg) based on the selling prices from 2007 to 2017 [28]. Historic prices for soy oil and soy meal are listed in Table B3 in the SI. Combining the unit costs of each product with their associated masses resulted in allocation factors of 0.45 and 0.55 of the market value produced as oil and animal feed, respectively.

Individual unit processes, such as algae cultivation and oil extraction, were simulated in Matlab®, and input and output values of the modeled system were used to calculate values of life cycle environmental impacts. All material and energy inputs and outputs associated with operation of the algal biorefinery were linked to subsequent harmful environmental impacts.

In addition, beneficial impacts associated with avoided emissions for conventional fuel and feed, replaced by algal co-products, were credited to the modeled system differently for each of the three functional units, using the system expansion method. For the functional unit of 1 MJ fuel, environmental impacts associated with conventional animal feed replaced by defatted algae (the coproduct) were credited to the system. The substituted animal feed consisted of a mixture of soy and corn used in conventional swine and poultry feed, of which up to 15 % can be replaced by defatted algae on a one-to-one basis [29,30]. For the functional unit of 1 kg animal feed, environmental impacts associated with each MJ of fossil diesel fuel substituted by algal biofuel (the co-product) were credited to the system. Additional details on the substitutions of feed and fuel are included in Section B of the SI. For the functional unit of 1 ha area, environmental impacts associated with both conventional animal feed and fossil fuel were credited to the system (see Figure 1).

### *Biorefinery*



case growth and composition parameters were obtained from Huntley et al. (2015) [31], and uncertainty ranges from Gerber et al. (2016) [5]. Algal productivity values used in the model were 14, 23, and 32  $\text{gm}^{-2}\text{day}^{-1}$  for minimum, median, and maximum values, respectively [5,31]. Lipid content values used in the model were 23, 37, and 51 percent, for minimum, median, and maximum values, respectively [5,31]. Algae were harvested after in-pond gravity settling, and dewatered using a belt filter press [24]. Daily energy and nutrient requirements for algae cultivation and dewatering are listed in Table A1 in the SI.

A representative “wet” solvent extraction process was modeled to illustrate the effect of the three functional units on LCA results for a multi-product biorefinery. The extraction process couples electromechanical pulsing to permeate algal cells, followed by oil separation with a membrane

[24,32,33]. The biocrude is recovered by distillation, and the defatted algae is dried prior to being used as an animal feed. Defatted algal biomass contains lipids in addition to carbohydrates and proteins, as the extraction process extracts 75% of the algal lipids leaving 25% of the lipid in the defatted residue [5,24]. Material and energy inputs and outputs for the oil-extraction process are listed in Table A2 in the SI [5].

## *2.2. Life Cycle Assessment*

The biorefinery was modeled with a cradle-to-gate boundary that included algae cultivation and biomass processing to crude oil and animal feed. On-site life cycle inventories for the base case are presented in Table B1 in the SI. In addition, environmental impacts associated with upstream processes, such as grid electricity and downstream processes, such as waste management of plastic, were accounted for, whereas emissions associated with product use were not included in the “well to gate” (or cradle to gate) framework. The LCA framework is described in more detail in Appendix B of the SI. The ecoinvent<sup>®</sup> v3.4 database was used for life cycle inventories of all upstream processes (e.g., grid electricity—for which we used the electricity mix in Texas) [34].

Ecoinvent v.3 has three types of models that can be incorporated into LCA studies: (1) cut-off, and (2) at the point of substitution (APOS), and (3) consequential. Differences among these three model types are described in detail in two papers [35,36]. Briefly, two of these models, cut-off and APOS, are intended to be used for attributional LCA. The difference between the two attributional models lies in the way they treat byproducts, i.e., waste materials, and waste materials that can be recycled. Cut-off allocates no environmental impacts to waste products and all impacts (even those associated with treatments of waste products that are recycled) are

attributed to the main product [31]. APOS, on the other hand, allocates impacts associated with required treatment processes of byproducts (i.e., waste that is disposed or recycled) to the byproducts [31]. The consequential models are intended to be used as background processes in consequential LCA studies. The consequential unit process models avoid allocation and use the system expansion method to account for products replaced by byproduct that substitute other products [31]. In addition, the consequential models account for expected changes in the marginal demand for each modeled product.

For this study, the cut-off model was used to model all upstream impacts, as the cut-off model type is the “descendant” of the default model in ecoinvent v.2. Although we used the system expansion method to handle coproducts, which is a consequential approach to LCA, we chose to use an attributional model for upstream processes. This choice was made to illustrate the consequential effects of our modeling strategy, and not the effect of the ecoinvent models. However, since the results of this study depend strongly on the results from ecoinvent, we present modeling results for the algal biorefinery using the APOS and Consequential models in Figures B2 and B3 in the SI. The data presented in Figure 3 and Figures B2 and B3 demonstrate that the comparative conclusions drawn from this study do not change with ecoinvent model type. This may not be true for other LCA models, and the effect of ecoinvent model type should be tested for individual case studies. The differences between the results for ecoinvent’s attributional models (i.e., Cut-off and APOS), which were very similar to each other, compared to ecoinvent’s consequential models are discussed elsewhere and are beyond the scope of the present study [35,36].

IMPACT 2002+ was used as the life cycle impact assessment method, because it allows us to calculate four synthetic damage categories (or endpoints): climate change (kg CO<sub>2</sub>-eq) (also a midpoint category), resource depletion (MJ of non-renewable energy), ecosystem quality (probability of disappearing fraction of species (PDF)•m<sup>2</sup>•year), and human health (disability adjusted life years (DALYs))[37]. Using four synthetic endpoint categories can help stakeholders interpret results, as it is easier to evaluate results for four endpoints than for seventeen midpoint categories that are characterized in IMPACT 2002+ and ReCiPe [37,38]. Since one goal of LCA, as an engineering design tool, is to improve the environmental performance of a product or process without shifting environmental burdens from one impact category to another (e.g., from climate change to ecosystem quality) [39], it is important to present tradeoffs among categories (such as climate change versus ecosystem quality) that can be more easily interpreted. Additional information on the impact categories for IMPACT 2002+ is included in Appendix B of the SI. After computing the four environmental indicators per each functional unit (e.g., kg CO<sub>2</sub>-eq per ha), indicator values were converted to “per 1 MJ” of fuel units (e.g., kg CO<sub>2</sub>-eq per MJ). This unit conversion was based on the amounts of fuel and animal feed produced, as well as the production area of the biorefinery. This was done to present all results in the same units, which allows the comparison of environmental indicators across functional units in one plot.

Since ISO 14040 and 14044 recommend the use of midpoint categories [7,8], and to ensure that the conclusions drawn from the results of this study are valid, we also calculated midpoint environmental impacts using the ReCiPe impact assessment method [38]. The results for

midpoint categories using ReCiPe, presented in Figures B4–B9 in the SI, agree with the results for the damage categories for IMPACT 2002+, which are presented in the main manuscript.

To account for parameter uncertainties, one thousand Monte Carlo simulations were run for each of the three models (each model has a different functional unit, as described above). This number of Monte Carlo simulations was shown to be sufficient for the present modeling framework previously [5]. Uncertainty ranges for foreground parameters (i.e., at the biorefinery) were taken from Geber et al. (2016) [5] and are listed in Tables C1 and C2 in the SI. For all upstream parameters, probability distribution functions were fit to outputs of 1000 Monte Carlo simulations run in the Simapro software package. The unit processes run in Simapro came from ecoinvent<sup>®</sup> version 3.4, and the parameters for each resulting distribution are reported in Table C4 in the SI. For the uncertainty analysis, normalization factors (reported in Table C3 in the SI) were applied to express each indicator in units of “points of environmental damage” (LCA points) to facilitate a comparison among the four damage categories [37]. One LCA point represents the impact of one person in one year in the United States. We used U.S.-specific normalization factors [40], because the algal biorefinery was assumed to be located in Texas. Since ISO advises against weighting [7,8], we did not sum the normalized values of the four damage categories to calculate an overall impact, but reported individual values for each damage category.

## **Results and Discussion**

When applying the system expansion method, the choice of functional unit—1 MJ of fuel, 1 kg of animal feed, or 1 ha of cultivation area—results in major differences in environmental impacts

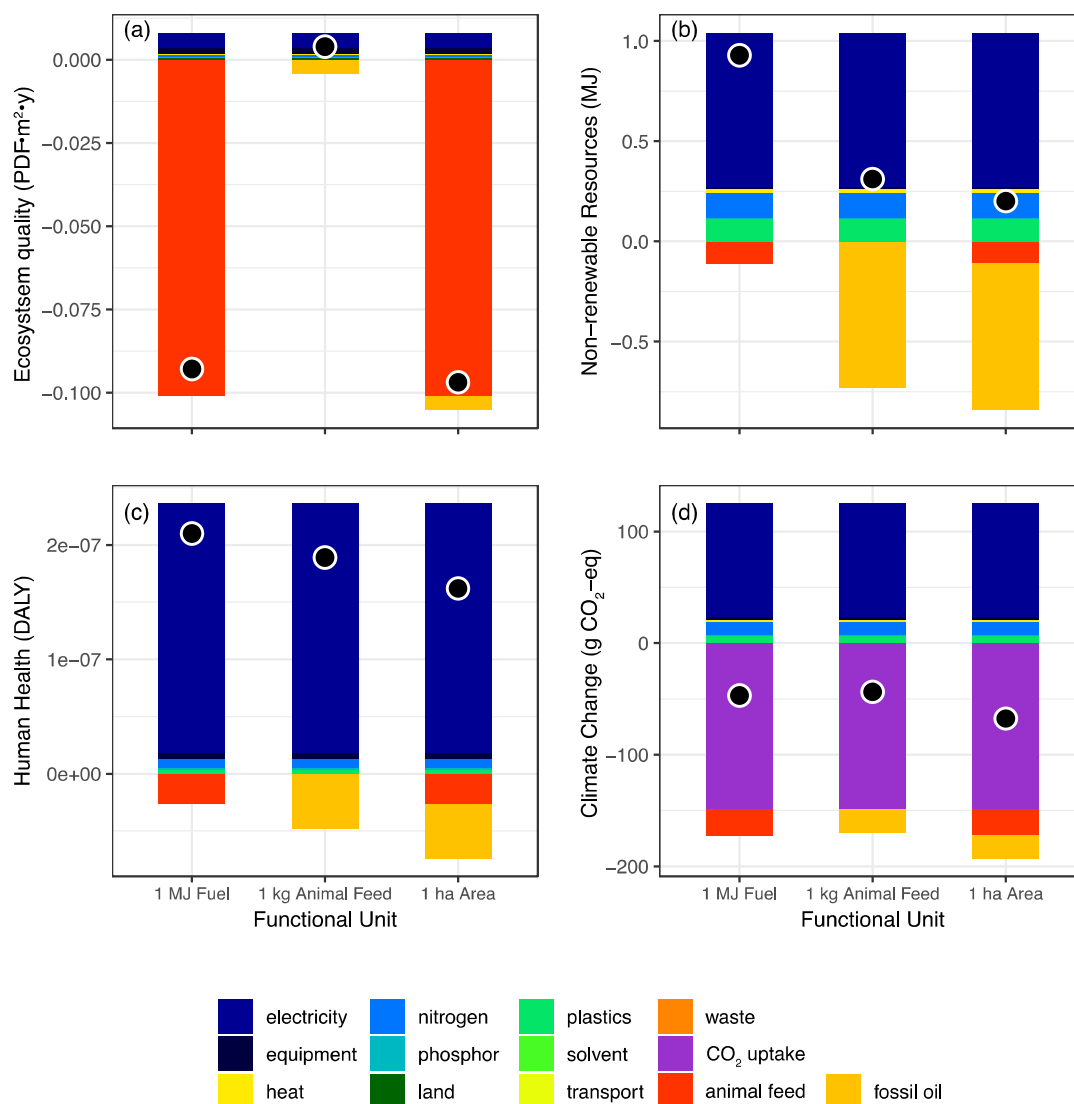
for two LCA indicators: ecosystem quality and non-renewable resources (Figure 3 (a), and (b)). Because environmental impacts are reported per 1 MJ of fuel produced, all harmful impacts (with positive magnitudes) are the same for the three functional units. The differences result from the way avoided impacts from co-products that displace conventional products (soy- and corn-based animal feed in red, and fossil fuel in yellow, Figure 3) are credited to the biorefinery using the system expansion method. When a functional unit of 1 kg of animal feed is used, the LCA model produces estimates of ecosystem quality impacts that are more harmful than for a model with a functional unit of 1 MJ of fuel or 1 ha of cultivation area (Figure 3 (a)). This results because avoided impacts for conventional feed (i.e., corn and soy) are credited to the two models that use a non-feed based functional unit (i.e., 1 MJ of fuel and 1 ha of area), but not to the system with a feed-based functional unit (i.e., 1 kg of animal feed).

The beneficial impact derived from replacing conventional animal feed with defatted algae results primarily from avoiding land use and fresh-water consumption to cultivate crops. Soy and corn use much more land than algae due to algae's high areal productivity, and algae can grow on non-arable land. Impacts associated with direct land use change for construction of algal raceway ponds—beyond those calculated with the two LCIA methods used in this study (IMPACT 2002+ and ReCiPe)—were not accounted for in the present modeling framework. Direct land use change impacts arising from transforming land to construct raceway ponds, however, have been shown to lead to life cycle climate change impacts [41].

The beneficial impact derived from replacing conventional fuel, which is credited to the two models that use a non-fuel based functional unit, is relatively small—i.e., 25 times less than the

impact for conventional animal feed. There is evidence in the literature that development of fossil fuels is impacting ecosystems [42]. More research should be conducted to estimate the available LCI data for elementary flows related to ecosystem impacts of fossil fuel production, which might be scarce. Most LCA studies of algal biorefineries focus on greenhouse gas emissions and energy balances. The analysis presented here, however, highlights the importance of evaluating environmental impacts for an indicator that assesses effects of algae on land use (i.e., the ecosystem quality indicator).

When using a functional unit of 1 MJ of fuel, impacts for depletion of non-renewable resources are more harmful (i.e., values with larger magnitudes) than for the other functional units, because avoided impacts of fossil fuels are credited to the two models that use a non-fuel functional unit (i.e., 1 kg of animal feed and 1 ha of area), but not to the system with a fuel-based functional unit (i.e., 1 MJ of fuel) (Figure 3 (b)). In summary, the model with the fuel-based functional unit benefits from avoided impacts associated with conventional animal feed, and the model with the feed-based functional unit benefits from avoided impacts associated with fossil fuel. The model with the land-area-based functional (1 ha area), on the other hand, treats the two biorefinery products (fuel and feed) equally. When a functional unit of 1 ha area is used, the model benefits from avoided impacts for both conventional products—fossil fuel, as well as soy- and corn-based animal feed.



**Figure 3.** Base case life cycle environmental impacts for four environmental indicators: (a) Ecosystem Quality, (b) Non-renewable Resources, (c) Human Health, and (d) Climate Change. Environmental indicators are reported in their representative units per 1 MJ of algal biofuel produced for the three functional units. The model boundary is “well to gate.” Upstream impacts were modeled using the cut-off unit process models in ecoinvent v.3.4. Colored bars represent impacts for individual unit processes, and black circles represent overall impacts. Values with positive magnitudes represent environmental harm and values with negative magnitudes represent environmental benefits.

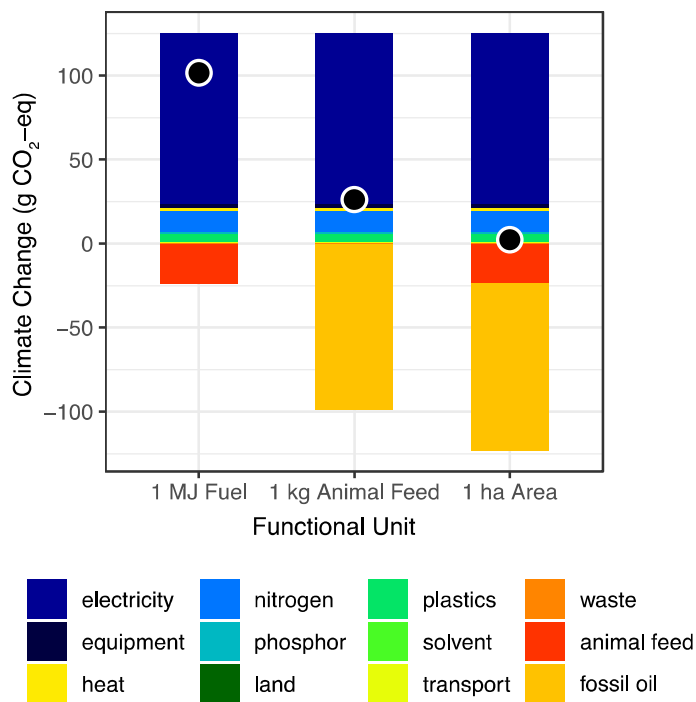
The choice of functional unit has less effect on the human health and climate change indicators (Figures 3 (c) and (d)) because these metrics are dominated by electricity and CO<sub>2</sub>, respectively.

For the human health indicator, impacts for the three functional units are of the same order of magnitude. Because a difference of at least three orders of magnitude is required to conclude that one process is more harmful than another with respect to human health [39], we conclude that differences shown in Figure 3(c) may not be meaningful. The impact of an algal biorefinery on human health is harmful regardless of functional unit choice. The avoided environmental impacts from fossil fuel and conventional animal feed, which are credited to the system, cannot overcome the harmful impacts on human health associated with electricity consumption at the algal biorefinery. Reducing power for pumping, mixing, and CO<sub>2</sub> delivery, or using renewable electricity can improve the environmental performance of algal biorefineries for all impact categories, including human health [5,24]. This large harmful effect of electricity consumption at algal biorefineries demonstrates the importance of presenting detailed LCA results that stakeholders can interpret.

For climate change, and a “well to gate” model boundary, the algal biorefinery is beneficial to the environment for the three functional units, because the carbon dioxide sequestered during algal growth offsets harmful emissions (Figure 3c). Using a functional unit of 1 ha area results in a climate change impact that is approximately 30 percent more beneficial compared to the other two functional units, because avoided impacts are credited the modeled biorefinery for both fuel and feed. This increased benefit results from the avoided impact from fossil fuel and conventional animal feed, which are credited to the biorefinery. For climate change, a difference of 10 percent is sufficient to claim that one system is more environmentally beneficial than another [39]. This difference, therefore, may be significant, but uncertainty analysis is needed

before drawing such a conclusion. In addition, this result prompted an analysis of the effect of model boundary for the climate change indicator.

The effect of model boundary on LCA results for the climate change indicator, using the system expansion method to handle co-products, are illustrated by comparing the results presented in Figure 4 to the results presented in Figure 3(d). If a “well to wheels” boundary is used instead of the “well to gate” boundary, carbon dioxide emissions from fuel combustion and animal feed digestion would negate the beneficial impact of carbon dioxide capture during algal growth. In other words, the carbon sequestered during algal growth would be released back to the atmosphere during the use phase of the products. LCA treats biogenic carbon that is sequestered (during biomass growth) and released during product use as net neutral. The carbon sequestered and emitted during the cultivation of algae and use of algal products, respectively, are therefore not shown in Figure 3. For an LCA model with a “well to wheels” boundary, however, avoided *fossil* CO<sub>2</sub> emissions from fossil fuel combustion are credited to the systems with non-fuel functional units, i.e., 1 kg of animal feed and 1 ha area (Figure 4). For the “well to wheel” boundary (Figure 4), the choice of functional unit affects the climate change indicator more than for the “well to gate” boundary (Figure 3(d)). For the “well to gate” boundary, the biorefinery is a net sequesterer of CO<sub>2</sub> for the three functional units (Figure 3c), whereas for the “well to wheels” boundary, the biorefinery is a net emitter of CO<sub>2</sub> for two functional units (1 MJ fuel and 1 kg animal feed) and is CO<sub>2</sub> neutral for the functional unit of 1 ha area (Figure 4). This demonstrates the importance for choice of model boundary (based on the goal of the study), as well as choosing the most appropriate functional unit.



**Figure 4.** Effect of functional unit on the climate change indicator using “well to wheels” boundary. Values with positive magnitudes represent environmental harm and values with negative magnitudes represent environmental benefits.

A functional unit of 1 MJ of fuel is often used to compare the greenhouse gas emissions of biofuel alternatives to fossil fuels. For example, LCAs that employ a “well to wheels” boundary coupled with a fuel-based functional unit are used to determine if biofuels are in compliance with the Renewable Fuel Standard (RFS). Figure 5 illustrates life cycle environmental impacts for a fuel based functional unit (1 MJ) and four methods used to handle co-products: the system expansion method, as well as allocation based on mass, energy, and market values of the fuel and animal feed produced by the algal biorefinery modeled here. The basis of allocation (mass, energy, and market value) results in different percentages of impacts distributed between fuel

(algal oil) and feed (defatted algae), which has been shown previously. Previous studies show the effect of co-product handling method on impacts of climate change for terrestrial biorefineries [18,20], but the data presented in Figure 5 show that the choice of co-product handling method can affect two environmental impacts (climate change and ecosystems) differently. For climate change, the system expansion method resulted in more harmful impacts than the three allocation methods, whereas for ecosystem quality the system expansion method resulted in more beneficial impacts compared to the three allocation methods. This is a result of the impacts that were credited to the system for the animal feed (or defatted algae) co-product for an LCA model that used the system expansion method. For climate change, the avoided impacts associated with conventional animal feed are not sufficient to overcome the harmful impacts of the algal biorefinery, which primarily result from the use of grid electricity. For ecosystem quality, however, the avoided impacts associated with conventional animal feed surpass the harmful impacts associated with the biorefinery.

An underlying assumption for allocation is that the material and energy inputs and outputs of the biorefinery and their associated environmental impacts can be distributed based on the mass, energy, or market value of the biorefinery products [20]. Allocation based on energy makes sense for biorefineries that produce energy products only (e.g., liquid fuel and electricity), and thus may not be appropriate for a biorefinery that produces bio-based chemicals [20]. However, since one characteristic of animal feed is its caloric value we included energy based allocation. Allocation based on market value can vary widely because of market volatility, and may be inappropriate for a developing technology, such as algae, for which there is no existing market. Since there is no existing market for algae, we used market values for soy oil and soy meal to

represent algal oil and meal (i.e., defatted algae) (Table B3 in the SI), but we acknowledge that compositions of the algal products are different from the composition of the soy products. Furthermore, although allocation of environmental impacts among biorefinery products is simple to apply, it assumes that all processing steps are associated with all biorefinery products, which may not be true. For example, one could argue that allocating a processing step associated with only one product (e.g., upgrading of fuel) among all biorefinery products does not make sense. One reason the system expansion method is a preferable method for handling co-products, is because it does not artificially divide up environmental impacts of product specific processes. Other authors have claimed that as long as there are similar market opportunities for all biorefinery products, the displacement method is preferred over product-based allocation of environmental impacts [20]. Recently it was reported that from a market perspective, algae can be developed as a feedstock for both fuel and feed [43], making the use of the system expansion method acceptable. Results presented in Figure 5 demonstrate the challenge of providing a single value to stakeholders that represents the environmental impact of a biofuel compared to a fossil fuel, when using 1 MJ of fuel as the co-product, and illustrate why ISO recommends that allocation be avoided when possible.

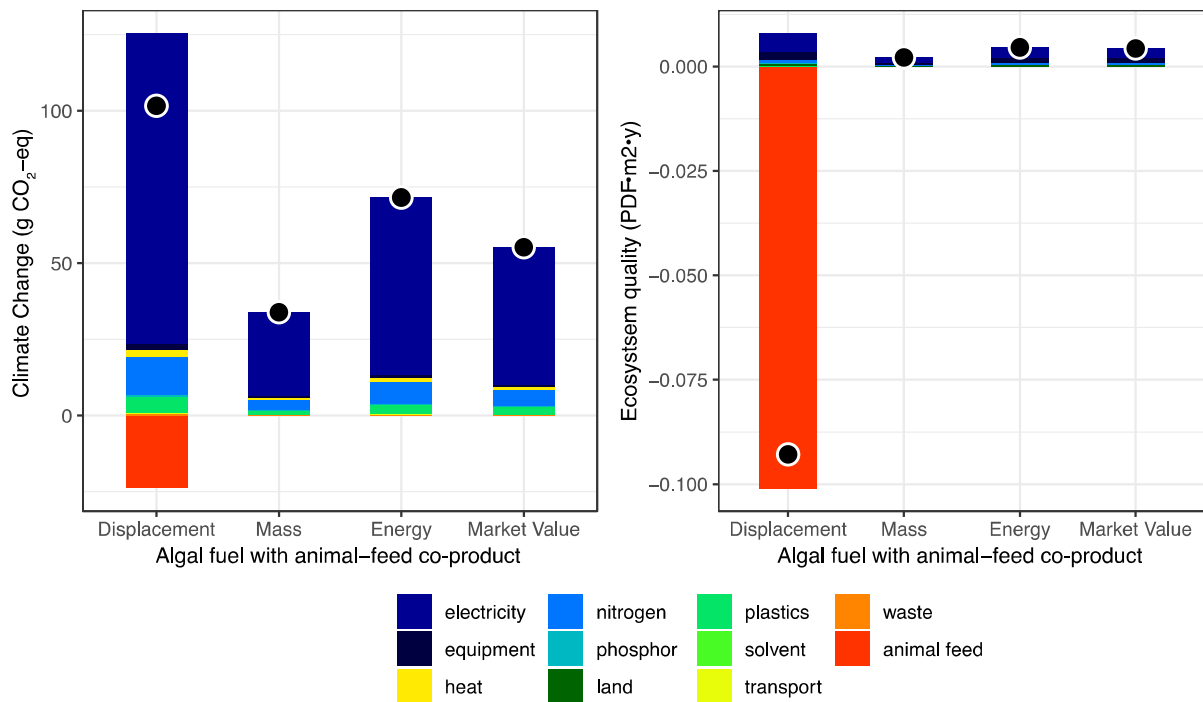
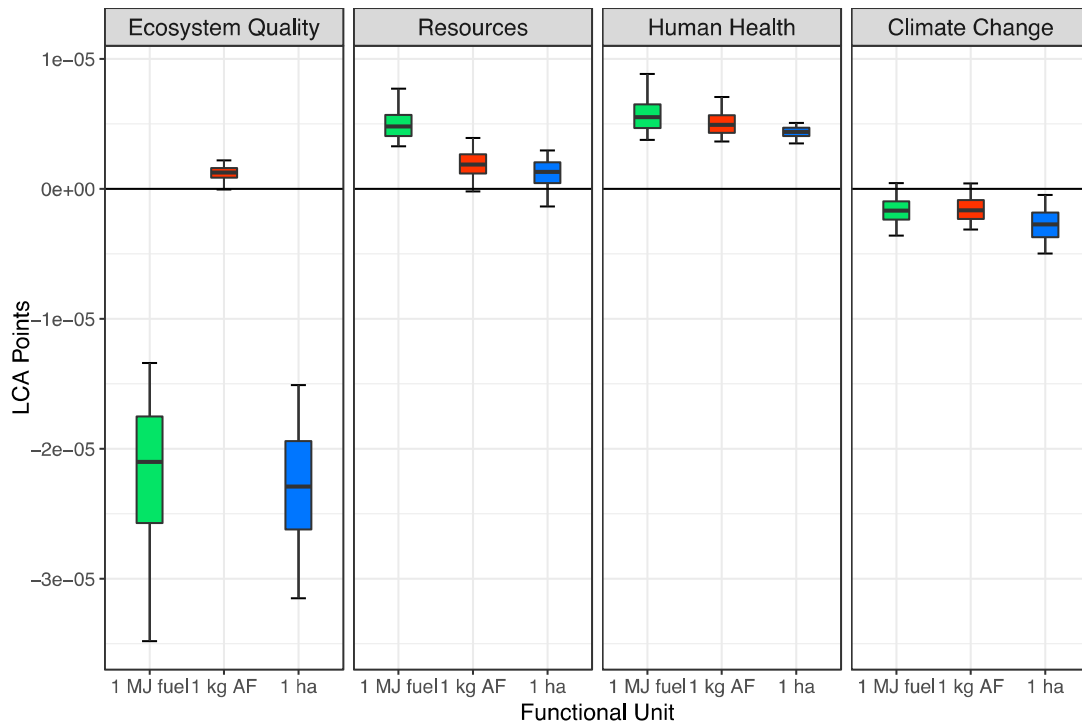


Figure 5. Effect of co-product handling methods on life cycle environmental impacts of climate change and ecosystem quality for 1 MJ of algal fuel, using the displacement (or system expansion) method, as well as allocation based on mass, energy, and market values of oil and defatted algae (to be used as animal feed).

Because cultivation of algae for fuel, feed, and other products is a developing technology without full-scale commercial data, presentation of data that accounts for parameter uncertainty is critical [2,5]. Alternative functional units can produce different environmental impacts for the ecosystem quality, and resource depletion indicators, even when parameter uncertainty is accounted for (Figure 6). For LCA models that use the system expansion method to handle co-products, normalized damage categories that account for uncertainty shows that for the fuel and land area functional units, the benefits for ecosystem quality are likely greater than the benefits for climate change (with a well to gate boundary) (Figure 6). For the animal feed functional unit, however, impacts are harmful for ecosystems and likely beneficial for climate change (Figure 6).



**Figure 6.** Environmental comparison of three functional units: 1 kg animal feed (AF), 1 MJ fuel, and 1 ha cultivation area with a “well to gate” model boundary. Impacts are expressed in unit of LCA points per 1 MJ of fuel produced for the three functional units: 1 MJ fuel, 1 kg animal feed (AF) and 1 ha area. Center lines represent median values, edges of boxes represent 25th and 75th percentiles, and limiting bars represent 5th and 95th percentiles of the distributions resulting from 1000 Monte Carlo simulations. Values with positive magnitudes represent environmental harm and values with negative magnitudes represent environmental benefits.

Additionally, a functional unit of 1 kg of animal feed results in a harmful impact for ecosystem quality, whereas functional units of 1 MJ of fuel and 1 ha of area result in a beneficial impact for the ecosystem quality indicator, even when accounting for the large uncertainties associated with modeling algal biorefineries (Figure 6). Similar to the base case results (Figure 3), the results for climate change and human health impacts that account for uncertainty did not change much with different functional units. This is illustrated by the overlapping distributions of impacts for the three functional units associated with the climate and human health indicators. However, for the

non-renewable resources indicator, functional units of 1 kg animal feed and 1ha area likely result in less harmful impacts compared to 1 MJ of fuel, due to beneficial impacts that result from avoided impacts for fossil fuel. Since LCA of algal biofuels is associated with large uncertainties [2,5], it is notable that the choice of functional unit may affect LCA results for two out of four environmental indicators modeled here, even when accounting for uncertainties.

For the displacement method, the choice of functional unit affects LCA results for multi-product algal biorefineries, because algal oil (used to produce fuel), and defatted algae (used to produce animal feed) both represent significant components of the biomass by mass and energy. This is a known issue for terrestrial biorefineries for cases in which multiple products make up significant fractions of the biomass [18]. The use of any co-product handling method was avoided in a study that demonstrated that algae could improve the sustainability of aquaculture study by using a functional unit of 1 kg of algae [13]. In this study, whole algae was compared to soy, a crop that also supplies oil and meal. For the scenario modeled by Beal et al., (2018), all of the algal biomass (i.e., oil and defatted biomass) were used as a feed product, and there was no need to distribute impacts among co-products. This is a unique case study for an algal biorefinery that produces a single product, which will not translate to most multi-product systems. For the present study, a model with a functional unit of 1 kg of algal biomass would produce similar overall results to those presented here for a model with a functional unit of 1 ha area. LCA models with both functional units (1 kg algal biomass and 1 ha area) include avoided impacts for all products (e.g., fuel and feed), which are credited to the biorefinery. We chose the area based functional unit, however, because most often, the goal of algae cultivation is to utilize land to produce energy, nutritional, and other useful products.

To avoid allocation among co-products, Barr and Landis used a functional unit of 1 metric ton of omega-3 fatty acids and all co-products (i.e., animal feed and fuel) to model an algal biorefinery that produced a high value nutraceutical (i.e., omega-3-fatty acids) [44]. This choice of functional unit allowed the modelers to avoid allocation, but it makes it difficult to compare algae-derived omega-3-fatty acids to conventional production of omega-3 fatty acids. To compare algae-derived and conventional production of omega-3-fatty acids, the authors included soy production (to be used as animal feed) as part of the conventional production process for omega-3-fatty acids. Soy production was added to the conventional production model so both systems produced equivalent amounts of animal feed [44]. This is a valid modeling approach, but it complicates interpretation of the model results by potential stakeholders. Furthermore, the specific product-based functional unit used by Barr and Landis creates challenges for comparing these results to those for an algal biorefinery that focuses on other nutritional products or liquid fuel.

Synergistic combinations of products are critical for successful commercialization of algal biorefineries [14,15]. To develop environmentally sustainable products, the algae industry needs to compare the environmental performance of biorefineries that produce different products, such as fuel, food, and nutraceuticals. Thus if the goal of an LCA is to evaluate environmental impacts of multi-product algal biorefineries to determine the best way to use algae, a functional unit should be crafted that considers all algal products. This agrees with the recommendation from ISO 14040 and 14044 and the ILCD Handbook, which state that when choosing a functional unit all functions of the production system should be considered [10,39]. *We therefore*

*propose that a functional unit of 1 ha area be used for LCA models with a goal of evaluating multi-product algal biorefineries.* For a functional unit of 1 ha area, the function of the system consists of the use of land, as an available resource, to cultivate algal biomass and produce energy and nutritional products (in the case of the present study). This functional unit could be expanded for algal biorefineries to include additional functions, e.g., to provide pharmaceuticals or commodity chemicals. Since we view algae production as a way to utilize land, we recommend this functional unit. Although the land-based functional unit results in less harmful environmental impacts compared to the two product based functional units modeled here, it is not our intention to produce favorable results for algal systems. We propose a land based functional unit, because it assesses impacts in an equitable manner, by accounting for avoided emissions associated with all products from the biorefinery. In future studies, the use of a land-based functional unit could facilitate accounting for direct and indirect land use change due to cultivation of algae for feed production. Development of an integrated assessment model framework, however, has advantages over LCA for assessing land use change impacts of algae [45].

Using a functional unit of 1 ha area combined with the system expansion method accounts for replacing conventional products, such as fossil fuel and conventional animal feeds. Since the algae industry is developing and not yet mature, it is important to emphasize potential improvements in environmental performance when using algae compared to conventional products. This approach accounts for the environmental effects of an emerging industry as it expands. However, if algal fuel and feed production mature into a full-scale commercial industry, the approach proposed here—i.e., crediting the algal biorefinery with avoided fossil

fuel, corn and soy productions—would not be appropriate. In that case, alternative modeling approaches (e.g., integrated assessment) could be used to represent energy and agricultural markets and their interactions with climate change impacts over time as the algae industry develops [45]. If the goal of an LCA is to compare the environmental impacts of algal fuel to other liquid fuels (e.g., fossil or terrestrial biofuels), however, a functional unit of 1 MJ of fuel or 1 km travelled (for a specific type of vehicle) is appropriate. LCA results, in this case, should also include an analysis of alternative methods for handling co-products.

### **3. Major Findings**

The correct choice of functional unit depends on the goal of an LCA applied to multi-product algal biorefineries. In addition, all methods used to handle co-products have limitations. If the goal of an LCA is to compare the environmental impacts of algal fuel to other liquid fuels (e.g., for regulatory purposes), a functional unit of 1 MJ fuel will likely be required. In this case, multiple co-product handling methods should be analyzed and presented to stakeholders. If the goal of the LCA is to compare among options in the design of a multi-product biorefinery, however, a functional unit that accounts equally for all products, such as an area based functional unit should be used in combination with the system expansion method. This modeling framework will help elucidate the environmental impacts of alternative algae cultivation technologies, conversion pathways, and algal products to determine the most sustainable use of algae. This will enable the assessment of products such as nutraceuticals needed for algae to be economically viable, along with fuel products that may be more desirable for curbing climate change.

## **Acknowledgements**

Funding was provided by the US Department of Energy (Grant: DE-EE0007091). We thank members of the Duke Marine Algae Industrialization Consortium (MAGIC) for their work that supported this manuscript. We thank Calvin Whealton, Paul Scherrer Institute, for help with computer code.

## **Author contributions**

D. Sills and L. Gerber Van Doren contributed to conception of the study. D. Sills, L. Gerber Van Doren and C. Beal contributed to analysis and interpretation of the data, drafting of the article, critical revision of the article, and final approval of the article. Elizabeth Raynor contributed to analysis and interpretation of the data, critical revision of the article, and final approval of the article. Deborah Sills (deborah.sills@bucknell.edu) takes responsibility for the integrity of the work as a whole from inception to finished article.

## **Statement of Informed Consent, Human/Animal Rights**

No conflicts, informed consent, human or animal rights are applicable.

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