Quantifying Variability in Life Cycle Greenhouse Gas Inventories of Alternative Middle Distillate Transportation Fuels

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Quantifying Variability in Life Cycle Greenhouse Gas Inventories of Alternative Middle Distillate Transportation Fuels

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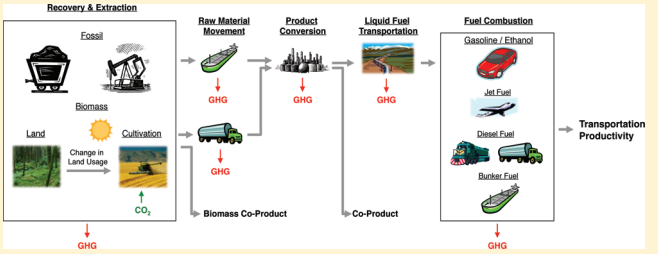
Supporting Information

ABSTRACT: The presence of variability in life cycle analysis (LCA) is inherent due to both inexact LCA procedures and variation of numerical inputs. Variability in LCA needs to be clearly distinguished from uncertainty. This paper uses specific examples from the production of diesel and jet fuels from 14 different feedstocks to demonstrate general trends in the types and magnitudes of variability present in life cycle greenhouse gas (LC-GHG) inventories of middle distillate fuels. Sources of variability have been categorized as pathway specific, coproduct usage and allocation, and land use change. The results of this research demonstrate that subjective choices such as coproduct usage and allocation methodology can be more important sources of variability in the LC-GHG inventory of a fuel option than the process and energy use of fuel production. Through the application of a consistent analysis methodology across all fuel options, the influence of these subjective biases is minimized, and the LC-GHG inventories for each feedstock-to-fuel option can be effectively compared and discussed. By considering the types and magnitudes of variability across multiple fuel pathways, it is evident that LCA results should be presented as a range instead of a point value. The policy implications of this are discussed.



1. INTRODUCTION

Variability, although inherent in life cycle analysis (LCA), is typically not explicitly considered. Instead, results are reported as a point value,[1−5] or when variability is addressed, it is often evaluated by comparing point values from multiple studies.[6,7] These approaches lack the ability to develop new data sets to target the sensitivity of specific factors, which could then be used to understand best practices for reducing LC-GHG emissions. In one notable exception, Farrell[8] examined variability in life cycle greenhouse gas (LC-GHG) inventories from corn ethanol by recreating the results of other studies and rectifying inconsistencies in metric choice and system boundaries. Such analyses have the potential to identify areas where improvement could reduce LC-GHG emissions for emerging fuels where facilities do not yet exist.

Delucchi[9] argue that LCA is a limited input-output representation of energy use and emissions that lacks the policy parameters or market functions needed to relate the results to policy actions. Indeed, an attributional LCA is a simplification of a complex system that is intimately linked to market effects. As a consequence of these simplifying assumptions, variability is introduced to LCA results that hinder comparisons of different fuel pathways.

To understand how variability impacts LC-GHG inventories of transportation fuels, a new methodological approach was developed using screening level LCAs. Screening level analyses provide preliminary assessments of technology alternatives with the intent of informing research funding and decision makers.[10] A requirement of screening level LCAs is to identify the pivotal factors defining the LC-GHG emission profiles of fuel production for each LC step and each feedstock. Optimistic, nominal, and pessimistic sets of these key parameters were developed to formulate corresponding low LC-GHG emissions, baseline or nominal LC-GHG emissions, and high LC-GHG emissions scenarios for each feedstock-to-fuel pathway; hence, results for each feedstock-to-fuel pathway are a range of possible LC-GHG inventories intended to demonstrate variability in fuel production processes.

This new methodological approach was used to develop LC-GHG inventories for a range of Synthetic Paraffinic Diesel (SPD) fuel pathways as well as conventional diesel fuel from conventional crude oil and Canadian oil sands. SPD is defined as hydrocarbon fuel with similar molecular composition to conventional diesel fuel but containing zero aromatic compounds and zero sulfur. This definition follows that of Synthetic Paraffinic Kerosene (SPK).[11] SPD and SPK are considered "drop-in" alternatives because they can serve as direct replacements for conventional fuels with little or no modification to existing infrastructure or vehicles. This work examines SPD fuels created from the gasification and Fischer−Tropsch synthesis of coal, natural gas, or biomass (switchgrass, corn stover, and forest residues) and the hydroprocessing of renewable oils (from soybeans, palm, rapeseed, algae, jatropha, and salicornia). In

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addition to the aforementioned fuel options, conventional diesel fuel from oil shale was also examined leading to a total of fourteen feedstock-to-fuel combinations.[12] While these encompass a wide range of replacement options for middle distillate fuels, it is not all-inclusive. Details regarding the recovery and processing of each feedstock are given in Table 1 of the Supporting Information. Although the results in this study are specific to middle distillate transportation fuels, these conclusions are applicable across all energy products.

This set of middle distillate life cycle (LC) greenhouse gas (GHG) inventories is used to quantitatively demonstrate general trends in the types and magnitudes of variability. It does so by systematically quantifying effects of specific parameters on the LC-GHG inventory of multiple fuel options within a consistent analysis methodology. The arguments for the work are developed as follows: Section 2 presents the analysis framework used to develop LC-GHG inventories and defines the types of variability within LC-GHG inventories; Section 3 gives case studies using the techniques of Section 2 with quantitative examples; Section 4 integrates the results to yield a comparison of LC-GHG inventories of all fuel options considered within this work; and Section 5 concludes with an overview of key conclusions and a discussion of policy implications.

2. ANALYSIS FRAMEWORK

An analysis framework that results in a range of LC-GHG emissions, as opposed to a point value was developed to demonstrate the influence of variability on LC-GHG inventories. Engineering judgment of the authors was used to identify key parameters with both variability and a considerable influence on the LC-GHG emissions. These were subsequently used to formulate low, baseline, and high LC-GHG emissions scenarios.[12] For example, parameters such as process efficiency or biomass feedstock yield can exert considerable influence on the LC-GHG emissions of the fuel pathway and their value a decade into the future could have considerable variability; hence, these parameters were varied as part of the three scenarios. Well-defined parameters with a large impact on the LC-GHG emissions (e.g., CO_2 emissions per unit of fuel combusted), and parameters with large variability but a small impact on the LC-GHG emissions (e.g., feedstock travel distance from source to refinery), were generally not examined.

Appropriate values for the key parameters were determined through literature review and consultation with relevant experts. In general, industry average rather than marginal values were sought. Variation of key parameter values across the three scenarios could arise from differences in time frame (e.g., historical data versus future projections), different feedstocks (e.g., bituminous coal versus sub-bituminous coal), different technologies, or changes in process design. While the upper and lower bounds of values found in the literature were generally used in the low and high emissions cases, baseline values were those which were deemed most likely, most frequently occurring, or were the average or midpoint of those from the literature.

The alternative jet fuel LC-GHG inventories of Stratton et al.[12] were leveraged for this work to generate diesel fuel pathways. The Supporting Information provides a comparison of diesel and jet fuel production processes. All analyses were carried out using the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation framework (versions 1.8b and 1.8a) and its supporting data, both developed and maintained by Argonne National Laboratory.[5] A simulation year of 2015 was chosen, and default GREET data were used in the analysis of the pathways, except where more recent data were obtained. Data from the open literature were utilized to build upon the existing GREET framework and derive LC-GHG inventories for feedstocks not currently available in GREET.

Variability in LCA must be distinguished from uncertainty. Variability is a dispersion of discrete results, each of which has been measured or calculated with an inherent uncertainty in the result. In the context of this effort, the sources of variability have been grouped into three categories: pathway-specific variability; coproduct usage and allocation; and land use change (LUC). Section 3 examines each of these categories with quantitative data, while the Supporting Information provides additional qualitative description.

3. CASE STUDIES

Variability, which is introduced into LC-GHG inventories through pathway-specific factors, coproduct usage and allocation assumptions, and LUC emissions, was quantified through the use of case studies. By identifying trends in how variability manifests itself across multiple pathways, equitable pathway comparisons were developed.

3.1. Pathway-Specific Variability. The premise of screening level analyses is that all key processes are included, but only key parameters with considerable influence on the results are examined in detail. These key parameters can be examined to ascertain the pathway-specific variability that is present in all fuel options. The production of Fischer−Tropsch diesel fuel from a combination of coal and biomass (coal and biomass to liquids, CBTL) was chosen to examine variability using this approach.

Both biomass and coal could be used as feedstock to a single F-T plant because they could be processed into F-T fuel using similar technologies. The analysis considered average US coal as feedstock mixed with switchgrass, corn stover, or forest residues at varying weight percentages. Before entering the gasifier, biomass must be preprocessed with a combination of thermal treatment and milling. Only configurations with carbon capture were examined. Due to the lack of experimental data on coal and biomass F-T process efficiencies, coal-to-liquid F-T (CTL) plant efficiencies were modified to account for the extra power consumption of biomass preprocessing and CO_2 compression for sequestration.

Local sensitivity analysis was conducted on feedstock type, potential for LC-GHG emissions from direct LUC, CTL process efficiency, biomass weight percentage, biomass preprocessing efficiency, CO_2 capture efficiency (percentage of CO_2 captured from the process), and CO_2 compression energy (see tornado diagram of Figure 1). Each parameter was varied with all others held at their baseline values with the impact on LC-GHG emissions quantified as a percent change from the baseline value. This analytical approach shows that feed biomass weight percentage has the dominant influence. Since this is an operational parameter, the emissions from CBTL facilities will be largely dictated by economic, as opposed to technological, limitations.

3.2. Coproduct Usage and Allocation Methodology. In addition to a primary fuel product, transportation fuel pathways often result in coproducts. In order to allocate emissions among products, a usage must first be defined for the coproduct. The coproduct usage may be well established, as is the case for the liquid fuels produced within an F-T product slate; however, this

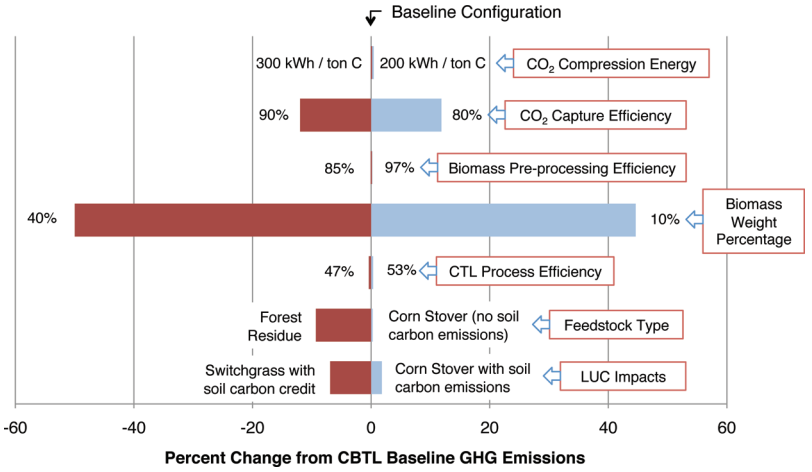


Figure 1. Pathway-specific sensitivity analysis of operational specifications and configurations of F-T diesel fuel from coal and biomass with carbon capture. Baseline LC-GHG inventory for CBTL fuels is 56 gCO_2e/MJ. Low, baseline, and high values for individual parameters were based on ranges from the literature and the engineering judgment of the authors.

is not always be the case, as with algae oil or jatropha oil where the biomass coproducts could have several different uses based on local economic conditions. In the absence of a specific configuration, the coproduct usage must be specified as part of the LCA. Four allocation methods were examined to assign LC-GHG emissions between the primary fuel product and any coproducts: mass allocation; energy allocation; market-value allocation; and displacement (a.k.a., system expansion). The Supporting Information contains a detailed description of these allocation methods.

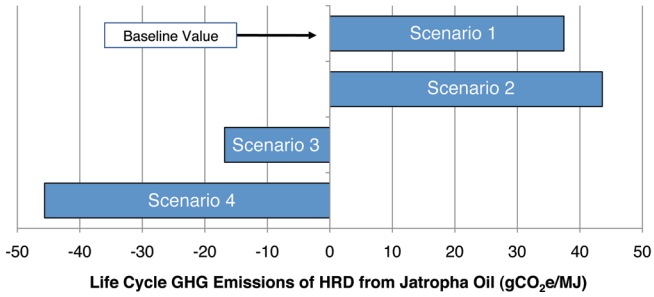
The choice of coproduct usage and allocation method may significantly affect the final results of the LCA. Several studies in the literature have acknowledged the variability introduced to LCA by different allocation methods.[6,10,12−15] Three examples were chosen herein to demonstrate the variability introduced by coproduct treatment: (1) the oil and biomass coproduct system of soybeans where the biomass has an existing market as an animal feed; (2) the oil and biomass coproduct system of jatropha capsules where the biomass coproducts have a variety of potential uses; and (3) the liquid fuel product slate from a coal and biomass fed F-T facility. All three examples show a general shortcoming in the displacement approach. When the coproduct creation is large relative to the primary product, the LC-GHG inventory of the primary product depends more strongly on the LC-GHG inventory of the displaced product than the processes and energy flows of the product being examined.

3.2.1. Fixed Coproduct Usage with Variable Allocation Methodology: Soy Oil. To examine liquid fuels created from soybean oil, the emissions associated with soybean cultivation, harvesting and oil extraction must first be allocated to the soy oil and meal. On average, a kilogram of soy oil is accompanied by 4.5 kg of soy meal, which has value as an animal feed. Allocation by mass, energy, and economic value were examined, as was system expansion wherein the soy meal displaced existing production of barley, corn, and soybean animal feed (equivalence was based on protein content). Details of the calculations are in the Supporting Information.

Using system expansion, the LC-GHG inventories can vary by 197.5 gCO_2e/MJ depending on the product displaced by additional soy meal production, from a low value of −159.5 gCO_2e/MJ with corn displacement to a high value of 38.0 gCO_2e/MJ with whole soybean displacement. By comparison, the LC-GHG inventory of conventional diesel is 90.0 gCO_2e/MJ. System expansion, as applied to oil and meal for animal feed, is inappropriate because of the large variation in LC-GHG emissions that would result from the choice of product displaced by soy meal (i.e., barley, corn, and whole soybeans). A more appropriate use of the displacement method would be achieved by modeling the resulting perturbation of incremental soy oil production on worldwide agricultural markets, but that is beyond the scope of a screening level, attributional, life cycle analysis. The displacement calculation is further complicated if LUC emissions are included for the soy meal, the displaced product, or both.

Depending on whether emissions are allocated based on mass, economic value, or energy, the LC-GHG emissions of soy oil based HRD were found to be 19.7 gCO_2e/MJ, 35.2 gCO_2e/MJ, and 28.9 gCO_2e/MJ, respectively. Allocation by mass would benefit a fuel producer, as it would result in most of the emissions being assigned to the meal instead of the oil. Furthermore, mass or energy allocation might not be appropriate as soy meal is valued based on its nutritional value, while HRD is valued based on its energetic content as a fuel. As such, market value allocation was adopted, for the purposes of this work, for all systems where oil extraction results in meal leaving the system for use as animal feed.

3.2.2. Variable Coproduct Usage with Variable Allocation Methodology: Jatropha Oil. On average, 1.7 kg of husk, 1.1 kg of shell, and 0.8 kg of meal accompany each kg of jatropha oil. The husks, shell, and meal could be burned for electricity or plowed into the earth to displace fertilizers. In addition, the meal could be sold as animal feed; however, detoxification is likely required because jatropha oil is mildly toxic to both livestock and humans.[16] Four scenarios of coproduct usage and corresponding allocation schemes were evaluated as shown in Figure 2. The combination of coproduct usage and allocation assumption was found to have a larger impact on the LC-GHG inventory of HRD from jatropha oil than the numerical inputs to the analysis. For example, scenarios 1 and 4 both assume the biomass coproducts are burned for electricity, but the different allocation methodologies result in more than a 200% change in the magnitude of the LC-GHG inventory. Scenarios 2 and 4 provide a second example



1	Co-product use:	Electricity
	Allocation:	Energy
2	Co-product use:	Fertilizer
	Allocation:	Displacement
3	Co-product use:	Animal feed, Electricity
	Allocation:	Economic value, Displacement
4	Co-product use:	Electricity
	Allocation:	Displacement

Figure 2. Sensitivity of LC-GHG emissions from jatropha oil HRD to coproduct usage and allocation assumptions. Baseline value indicates the chosen combination to represent HRD production from jatropha oil. Single usage and allocation entries indicate uniform application across all coproducts. Scenario 3 assumes meal is detoxified and used for animal feed with allocation by economic value, while all other coproducts are used for electricity with allocation by displacement of average grid electricity.

as they both adopt the displacement method, but the different coproduct usages of fertilizer creation or power generation again results in a 200% change in the magnitude of the LC-GHG inventory.

Unlike the case of soybeans, there is no process of elimination that can be used to determine a logical combination of coproduct usage and allocation methodology for jatropha. Since average analyses are not specific to any single production scheme, the decision should be made under the constraint of maximizing consistency between this fuel pathway and others where similar situations arise. This is a fundamental aspect of LCA that is often overlooked when fuel options are considered independently. Useful comparisons among fuel pathways can only be drawn through the application of consistent assumption sets. This is the second example, of what could be many, where subjective coproduct usage and allocation assumptions made by the LC analyst are as important than the underlying data.

3.2.3. Variable Process Configuration with Variable Allocation Methodology: F-T Fuel Production. Although diesel and jet fuel are the primary products of interest for an F-T fuel producer, naphtha is also created during wax upgrading. Since other F-T fuels are made as a result of producing either F-T diesel or jet fuel, the emissions from processing and all other upstream activities must be allocated among the fuels being produced. As previously emphasized, emissions allocation requires both a coproduct usage and methodology. In this analysis, F-T jet fuel is used by the jet fuel market, and naphtha serves as feedstock for high-octane gasoline via catalytic reforming; hence, both are utilized for their calorific value.

An F-T facility operator has some degree of control over the product slate of diesel, jet, and naphtha that are being produced. Figure 3 shows how a variable product slate affects the baseline LC-GHG inventory of F-T diesel from coal and switchgrass under an energy allocation scheme and a displacement scheme. When diesel is the dominant product (>70%), the displacement method produces similar results to energy allocation, and the two allocation methods yield the same result under a hypothetical product slate of 100% diesel. Reducing the fraction of diesel to the point where it no longer dominates the product slate causes the LC-GHG inventory calculated using the displacement method to diverge from the constant LC-GHG inventory calculated using energy allocation. When the yield of diesel is 75% by volume, there are 0.33 L of other F-T fuels produced for every liter of diesel. When the yield of diesel is reduced to 25% by volume, there are 3 L of other F-T fuels produced for every liter

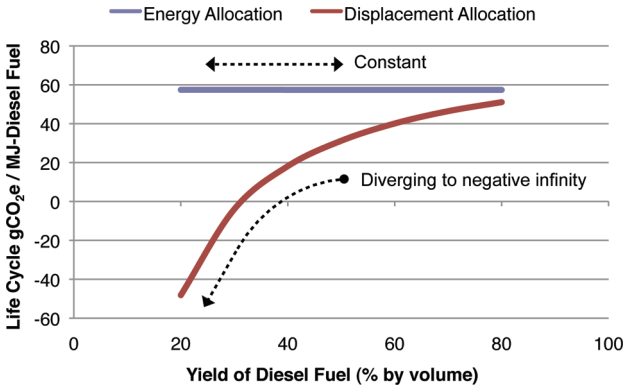


Figure 3. Influence of product slate composition on the LC-GHG inventory of F-T diesel fuel. Nondiesel fraction of product slate is composed of F-T jet fuel and F-T naphtha. Results are insensitive to the division of naphtha and jet fuel. Naphtha displacement follows procedure of Tarka[3] using petroleum fuel LC-GHG inventories of Skone and Gerdes.[1].

of diesel. In the limit where the percentage of diesel in the product slate approaches zero, the quantity of other F-T fuels produced per unit of F-T diesel asymptotically approaches infinity. If the other F-T fuels represent a reduction in emissions as compared to their petroleum equivalents, then the displacement method attributes these emissions reductions to the diesel fuel; therefore, the displacement methodology results in a diesel fuel which appears to have LC emissions that approach negative infinity as the yield is reduced toward zero. These trends are true for an LCA focused on jet fuel or naphtha; in fact, the divergence would be more substantial for jet fuel as it would, in reality, represent a smaller fraction of the product slate than diesel[12]

Using energy allocation among transportation fuel coproducts yields consistent results by preventing results from being skewed by subjective choices such as the selected product slate. The results using energy allocation come with the caveat that other fuels produced, such as jet fuel and naphtha, also affect the overall environmental impact of a particular feedstock-to-fuel pathway. Other coproduct usages for jet fuel and naphtha would change the product slate, allocation methodology, or both. For example, naphtha could be consumed internally for hydrogen production in which case it would not be a coproduct and it would be allocated zero emissions; further, naphtha could be used as a chemical feedstock in which case market-value allocation may be

more appropriate. These findings are also applicable to hydroprocessing of renewable oils, which can yield a product slate of diesel fuel, jet fuel, and naphtha.

3.3. Land Use Change (LUC) Emissions. Converting native habitats to cropland results in CO_2 emissions from organic carbon stored in plant biomass and soils. After fire, or another method, is used to clear the land and the leaves and fine roots have decomposed, a prolonged period of GHG release occurs as coarse roots, branches, and wood products decay or burn.[17] Conversely, crop cultivation on land with degraded or naturally low carbon stock could lead to net long-term carbon storage under proper future management practices. Because GHG emissions from LUC could dominate a LC-GHG inventory, LUC variability must also be quantified to understand LC-GHG inventory variability. The sign and magnitude of LUC emissions can vary substantially for any given feedstock depending on the prior land use, production and management practices, temporal considerations, and environmental conditions. Combining LUC emissions with feedstock yield adds further variability to the contribution of LUC to a LC GHG inventory since both are typically quantified per unit area of land.

Only GHG emissions from direct LUC, where land is converted to facilitate biofuel production, were examined in this study. No attempt was made to quantify the magnitude of indirect LUC emissions resulting from fluctuations in supply and demand for other agricultural products because of increased biofuel production. In order to properly capture these effects, detailed economic modeling is required which falls beyond the scope of this study (e.g., refs 4 and 18). From a qualitative viewpoint, indirect land use change emissions are simply the aggregated direct land use change resulting from increased prices for agricultural products; therefore, an appropriately chosen range of direct LUC emissions should bracket any potential indirect LUC.

This work used independent sets of results for each pathway under select LUC scenarios to account for the variability of if and when a fuel pathway may be subject to a particular type of LUC. Specifically, LUC was included for F-T diesel from switchgrass with and without cofeeding of coal,[19,20] soy oil HRD,[17] palm oil HRD,[17,21] rapeseed oil HRD,[22] and salicornia oil HRD.[23,24] In all scenarios, total LUC emissions were amortized over 30 years with no discounting. Palm oil HRD and F-T diesel from switchgrass are considered in greater detail as case studies to examine the influence of variability on the magnitude and sign of LUC emissions. The goal of this approach is to provide an understanding of how LUC emissions compare to the emissions from the other LC stages. It is not intended to explicitly quantify the specific LUC emissions that would result from the production of the feedstocks examined herein.

Four palm oil LUC scenarios have been examined in Figure 4 to show that LUC emissions are the dominant factor in the LC-GHG inventory of HRD from palm oils. The first scenario assumed no LUC emissions (denoted P0). The second scenario (P1) assumed direct LUC emissions from the conversion of previously logged forest to palm plantations.[21] It assumes that the LUC emissions from logging are not included in the system boundary. The third and fourth scenarios assumed LUC emissions resulting from the conversion of tropical rainforest (P2) and peatland rainforest in Southeast Asia (P3), respectively, to palm plantations.[17] Data for P1 were calculated from results presented in ref 21, while data for P2 and P3 were taken directly from ref 17. LUC data were corrected where appropriate for

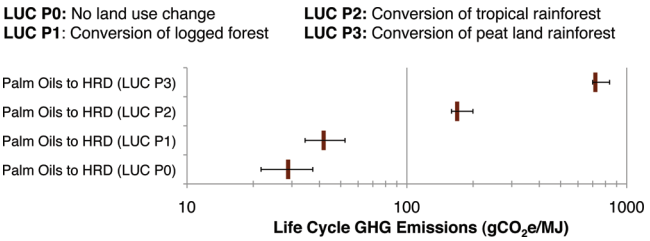


Figure 4. LC-GHG emissions of HRD from palm oil under four different land use change scenarios. Bar is baseline scenario, while whiskers correspond to low and high emissions scenarios. Horizontal axis is log scale so range spanned by low and high emissions scenarios is distorted differently for each LUC scenario.

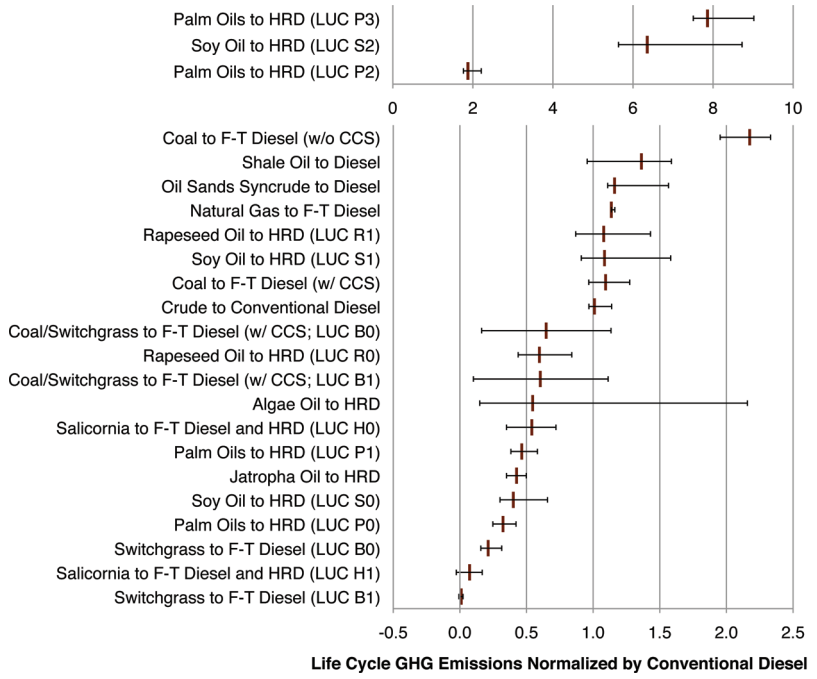
cultivation yield, time horizon, and coproduct treatment used in this work. If LUC emissions are not incurred (P0), palm oil HRD was found to have baseline LC-GHG emissions of 28.2 gCO_2e/MJ; however, it could increase to 705 gCO_2/MJ if the palm oil came from a palm plantation located on converted peatland rainforest (P4). Palm oil LUC can also be as small as 12.7 gCO_2e/MJ if the palm plantation is located on previously logged over forest and the LUC emissions for the logging is not included in the system boundary (P1). Furthermore, the potential variation in palm oil yield per unit land area causes the LUC emissions to vary by 121 gCO_2/MJ about the baseline value for the converted peatland rainforest LUC scenario (P4).

Two switchgrass LUC scenarios were examined. The first assumes no LUC (S0), while the second examines the potential for long-term soil carbon sequestration (S1). The latter scenario can occur where agricultural practices have led to a progressive and historical decline in soil carbon stocks[20] and the growth of switchgrass leads to conversion of carbon depleted soils to grasslands.[19,20] The estimate for soil carbon sequestration adopted herein is based on DAYCENT biogeochemistry model simulations from ref 19, who assessed soil LC-GHG fluxes and crop yields for switchgrass grown in Pennsylvania. These results are consistent with the range given by ref 20. When no change in soil carbon occurs, Fischer−Tropsch diesel made from switchgrass has a baseline LC-GHG emissions inventory of 18.2 gCO_2e/MJ. Sequestering carbon through switchgrass cultivation on carbon-depleted soils decreases this value by −19.8 gCO_2/MJ, leading to a potentially carbon negative fuel. If switchgrass is cofed with coal, which causes the baseline LC-GHG inventory to increase, then the magnitude of soil carbon sequestration per MJ will decrease.

Estimates of LC-GHG emissions from LUC are subject to both variability and uncertainty; however, uncertainty has not been considered here. This is consistent with the objective of providing an understanding of how LUC emissions compare to the emissions from the other LC stages. The variability introduced into the LC-GHG inventory of fuels subject to LUC has been emphasized because of the range and potentially dominant magnitude of prospective scenarios. These results demonstrate the importance of knowing the fuel feedstock origin when assessing how LUC emissions affect the environmental feasibility of a fuel option.

4. LIFE CYCLE GHG INVENTORY RESULTS

The life cycle analysis framework of Section 2 was used with the lessons from Section 3 to create Figure 5, which presents the LC-GHG inventories for a wide range of feedstock-to-diesel



Land Use Change Scenarios

Land use change	LUC Scenario 1	LUC Scenario 2	LUC Scenario 3
Switchgrass (B0, B1)	Carbon depleted soils converted to switchgrass cultivation	n/a	n/a
Soy oil (S0, S1, S2)	Grassland converted to soybean cultivation	Tropical rainforest converted to soybean cultivation	n/a
Palm oil (P0, P1, P2, P3)	Prev. logged over forest converted to palm plantation	Tropical rainforest converted to palm plantation	Peat land rainforest converted to palm plantation
Rapeseed oil (R0, R1)	Set-aside land converted to rapeseed cultivation	n/a	n/a
Salicornia (H0, H1)	Desert land converted to salicornia cultivation	n/a	n/a

Note: In all cases, LUC scenario 0 denotes no land use change

Figure 5. LC-GHG emissions for the alternative diesel fuel pathways under consideration. Uncertainty bars represent the variability captured by the low emissions, baseline, and high emissions scenarios. Please note the different scales for the top and bottom portions of the figure. Note: CCS denotes Carbon Capture and Sequestration although only the initial CO_2 compression energy is included; Land Use Change (LUC) scenarios are defined in Table S6 of the Supporting Information.

fuel pathways. Because of the uniformity of methodology, comparisons among pathways in Figure 5 are minimally influenced by subjective biases, and the range in LC-GHG inventory of each pathway is driven by pathway-specific variability and changes in coproduct assumptions necessitated by changes in process configurations. The data in Figure 5 have been normalized by the LC-GHG emissions of conventional diesel fuel to reflect their purpose as a cross-comparison of alternative diesel fuel options. The uncertainty bars in Figure 5 show the variability that is represented by the range of emissions as given by the low and high emissions scenarios. The Supporting Information contains results for the baseline scenario of all fuel pathways broken out by LC step to allow for the identification of critical steps driving the LC-GHG inventory of each fuel pathway. The impact of LUC is captured through the use of multiple LUC scenarios, as was discussed in Section 3.3, and summarized within Figure 5.

Conventional petroleum was found to have the lowest LC-GHG emissions of any fossil-based diesel fuel pathway examined herein; however, considerable variability in the LC-GHG profile of conventional diesel occurs as a result of different crude oil extraction practices, transportation profiles, and refining efficiencies. As an example, crude oil from Canadian oil sands using asphaltene gasification to supply hydrogen for bitumen upgrading produces a marginal barrel of conventional diesel with 55% more LC-GHG emissions than the average barrel. Even conventional crude oil imported from countries such as Nigeria, whose GHG emissions from crude oil extraction are the highest of all major crude oil exporters to the US, can result in 12% higher LC-GHG emissions than the average barrel. With more intensive carbon capture on F-T facilities, some fossil-based fuels could offer marginal reductions in LC-GHG emissions compared to the average barrel of petroleum; however, the reductions are likely to be sufficiently small that the fuel production would be motivated by energy diversity concerns rather than climate change mitigation.

Few biofuels were identified with zero LC-GHG emissions, which is partially due to not using the displacement method to allocate emissions. Algae has the largest variability of all fuel options examined due to the numerous options for extracting the algal oil from the aqueous algae and the potentially large emissions associated with concentrating CO_2 to stimulate high growth rate algal cultures. The range in LC-GHG emissions of all fuel options examined was from 0 to 9.0 times those of conventional diesel. The majority of this range stems from considering multiple LUC scenarios for HRD from soy oil and palm oil. Only minor differences were found to exist in the LC-GHG inventories of diesel fuel compared to jet fuel from the same feedstocks.

5. DISCUSSION

All of the biofuel options examined in this work could potentially be produced with lower LC-GHG emissions than conventional diesel; however, the production of these same fuels could also have LC-GHG emissions that exceed those of conventional diesel. The difference is due to the LC-GHG intensity of the processes and the emissions resulting from LUC. For this reason, it is critical to emphasize that the use of renewable resources as feedstock does not guarantee an environmentally beneficial fuel. Knowledge of specific production details is required for any definitive conclusions to be drawn. This constitutes a strong argument for LC-GHG inventories of transportation fuels to be presented as a range.

LC-GHG emissions are only one of many considerations that need to be examined when evaluating the feasibility and sustainability of an alternative fuel option. These include, but are not

limited to, the environmental impacts on global climate change and air quality, the efficient usage of fresh water and land resources, and the economic cost of fuel production. While quantifiable comparisons that incorporate these other attributes is ideal, this research has demonstrated the challenges of assessing and comparing different fuel options using only a single attribute, LC-GHG emissions. Within the LC-GHG scope, this work has built a quantitative sense of the challenges of drawing useful comparisons between attributional LCA results. Three key conclusions can be drawn from the potentially dominating influence of variability due to coproduct usage and allocation and LUC assumptions: 1) minimizing variability across LCA results by maximizing methodological consistency is essential to making useful comparisons between fuel options; 2) the absolute result from attributional LCA's have a diluted physical meaning and are most effectively used as a comparative tool, given the condition from the first key conclusion; and 3) it is paramount that decision makers and the general public be given the range of LC-GHG emissions that could result from the production and use of these fuels. Such an approach emphasizes the importance of understanding the key aspects that determine the LC-GHG emissions from fuel production and use. Furthermore, it can do so before any production actually occurs. Such knowledge would help to develop technologies and policies that diversify our energy supplies and stimulate economic development while mitigating the LC-GHG emissions from transportation.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information. List of all fuel options considered as part of this research, an overview of modifications to fuel production processes for examining diesel fuel as opposed to jet fuel, details of allocation calculations for the soy oil and soy meal system, and a summary of results for the baseline scenario of all fuel pathways broken out by LC step. This material is available free of charge via the Internet at http://pubs.acs.org.

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