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Life Cycle Carbon Footprint of Linear Alkylbenzenesulfonate from Coconut Oil, Palm Kernel Oil, and Petroleum-Based Paraffins

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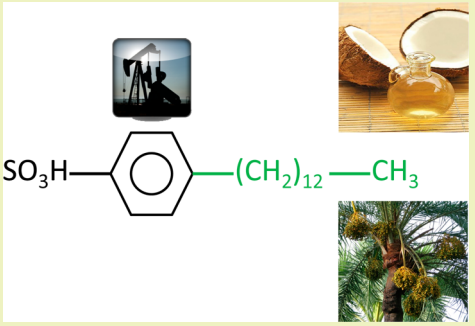
Supporting Information

ABSTRACT: Beyond renewable liquid transportation fuels from biomass feedstocks, there is great interest to use renewable feedstocks for sustainable chemical production. The goal of this study is to conduct an original cradle-to-grave carbon footprint for linear alkylbenzenesulfonate (LAS) using petroleum, coconut oil, and palm kernel oil derived paraffins, and including end of life emissions during wastewater treatment. The fully petroleum pathway to LAS was modeled using new inputs from industry (UOP). Mass allocation and energy allocation methodologies were both examined. We found that the greenhouse gas emissions from the production of LAS can be reduced between 45 and 50% by replacing petroleum based paraffins with coconut or palm kernel oil derived paraffins. End of life emissions of CO_2 during biodegradation in wastewater treatment was a major contributor to and differentiator of emissions in the carbon footprint. Direct land use change (dLUC) emissions of CO_2 were modeled using the IPCC method and were found to decrease life cycle emissions by at most 12% when either grasslands or shrublands are converted to either coconut or palm plantations.



KEYWORDS: *Linear alkybenzene (LAB), Linear alkylbenzenesulfonate (LAS), Renewable surfactants, Coconut oil, Palm kernel oil, Land use change*

INTRODUCTION

Literature Review. Linear alkylbenzene (LAB), commercially known as detergent alkylate, is one of the primary surfactant raw materials for the production of household laundry, dishwashing, and other surfactants.[1] The sulfonated form of this molecule, linear alkylbenzenesulfonate or LAS, is one of the largest volume commodity surfactants used today. The combination of good foam and detergency performance at low cost makes it very useful in many types of formulations.

Supported by UOP technology for large scale LAB production, LAS was introduced in the 1960s as a biodegradable alternative to sodium dodecyl benzenesulfonate (SDBS).[2,3] The global demand for LAB has since grown to more than three million metric tons per year with nearly 98% of LAB production being consumed in the manufacture LAS.[4] The technology for LAB and LAS production continues to evolve to become safer and more efficient. For example, the majority of LAB plants built since 1995 utilize solid bed catalyst technology instead of hydrofluoric acid to alkylate linear paraffin-derived olefins with benzene.

LAS is one of many high production volume chemicals in the market today and as such should be a high priority for improving its environmental performance.[5] As the chemical industry in the United States (and globally) moves toward more sustainable practices, an important part of this transition is greater utilization of renewable resources, such as biomass.[6] Planning for more sustainable solutions, the LAS technology is now further evolving to allow replacement of petroleum-derived linear paraffins with biorenewable sources of linear paraffins. The latter can be substituted without affecting downstream linear olefin and benzene alkylation steps of the commercial LAB and LAS manufacturing processes. However, biorenewable resources have environmental impacts associated with cultivation, transport, and conversion, which should be considered before any decisions are made to replace conventional fossil resources. Environmental life cycle assessment (LCA) is an appropriate methodology to evaluate environmental impacts in a comprehensive manner for product systems.

The authors of this study conducted a review of the current literature on environmental life cycle assessments for surfactants, and we are not aware of another study that developed a LCA for LAS production using renewable paraffins. Berna et al. reported an LCA for petroleum LAS production but did not include end of life (EoL) processes or their emissions.[7] The fate of petroleum LAS at its EoL (e.g., wastewater treatment) was also studied previously by Berna et

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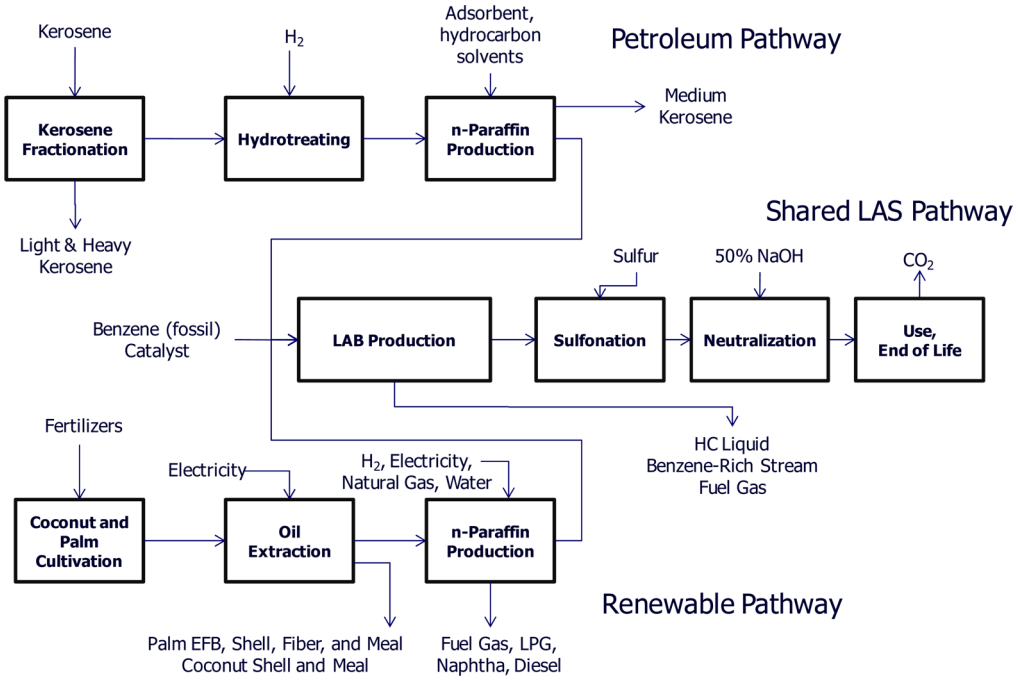

Figure 1. Product pathway diagram for petroleum and renewable LAS.

al., and we assume that renewable LAS degrades identically to petroleum LAS for the study reported here.[8]

The research objectives of this study are to (i) conduct a life cycle carbon footprint (LCCF) of petroleum LAS using updated LAS production inputs provided by UOP LLC, (ii) conduct an original LCCF of renewable LAS in which the paraffin group of LAS is from coconut oil (CO) or palm kernel oil (PKO) and the benzene is from petroleum, (iii) include in the LCA emissions of greenhouse gases (GHGs) generated during end of life (EoL) wastewater treatment, and (iv) estimate the magnitude of direct land use change emissions of GHGs due to cultivation of coconut and palm in the Asia–Pacific region of the world using an IPCC method.

Savings of GHG emissions compared to petroleum LAS are reported for coconut oil (CO) and palm kernel oil (PKO) LAS. This study will also include scenario analyses where energy allocation, rather than mass allocation, is applied; uncertainties in EoL emissions of LAS are studied; and comparisons are made between LAS with petroleum and renewable fatty alcohol sulfates (FAS), an alternative detergent to LAS.

LCA METHODS

Goal, Scope, and Functional Unit. The goal of this study is to calculate the cradle-to-grave GHG emissions from the production and disposal of fossil and renewable LAS, but use of LAS (including packaging) has been omitted as it is assumed to be the same for both renewable and petroleum production pathways. The system boundaries of the analyses are consistent with attributional LCA where only inputs, outputs, and emissions directly connected to the LAS pathway are included. CO (India) and PKO (Malaysia) are examined as renewable feedstocks for LAS as they are primarily composed of carbon chain lengths averaging 13–14, respectively, which are ideal for the production of LAS.[9] The functional unit for this study is 1 kg of surfactant, and we assume identical performance of petroleum and renewable LAS because the molecular structures of the fossil and renewable LAS are essentially identical.[10] We assume that sustainability principles, such as those proposed by the Roundtable on Sustainable Biomaterials (RSB) will be adhered to during commercialization of this new product so as to avoid competition with food and indirect land use change emissions.[11]

The LAS pathways for this study are shown in Figure 1, and major inputs and coproducts are shown for each process stage. Petroleum-based LAS begins with crude oil extraction (not shown in Figure 1) and a subsequent distillation of the kerosene fraction of the crude oil produces light and heavy kerosene as coproducts. The medium kerosene product is then hydrotreated before the linear (*n*-)paraffin fraction is recovered by means of a simulated moving solid adsorbent bed. Light hydrocarbon solvents are employed to produce high purity *n*-paraffin and a coproduct containing the non *n*-paraffin fraction of the medium kerosene. A small quantity of these solvents is consumed in the process. LAB production from the normal paraffins requires conversion of the linear paraffin to a linear olefin, which in turn is used to alkylate benzene with the linear olefin. The benzene is assumed to come from a fossil source. Several coproducts, including a benzene-rich liquid stream, hydrogen-rich fuel gas, and hydrocarbon (HC) liquid stream, are recovered from the LAB production stage. Sulfonation and neutralization require sulfur and an aqueous sodium hydroxide (NaOH) solution to produce a commercial grade 50% LAS solution. The EoL stage accounts for the emissions from the biodegradation of LAS to CO_2 in wastewater treatment plants (WWTPs) after its useful life. LAS derived from CO- and PKO-derived paraffins starts with cultivation of coconuts and palm fresh fruit bunches, respectively. Oil must then be extracted from the harvested materials, which is accomplished using a mechanical press.[12−14] The extraction process produces coconut meal and palm kernel meal in addition to the oils. Converting these biorenewable oils to normal paraffins requires a catalytic deoxygenation process that consumes hydrogen, natural gas, water, and electricity and generates coproducts of renewable fuels: diesel, fuel gas, LPG, and naphtha.[15] The

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processes that convert the *n*-paraffins to LAB are the same for both sources of *n*-paraffins, with EoL CO_2 emissions handled differently due to the source of the carbon in the LAS. In our study, fossil carbons will be counted toward the GHG total, while renewable carbon will not count, assuming carbon neutrality (CO_2 is sequestered into biomass during cultivation and then returned to the atmosphere during biodegradation in WWTPs). However, later in this article, we will consider changes in carbon stocks of the land cultivated for CO and PKO production (direct land use change emissions) and include those CO_2 emissions in the LCA.

Pathway Inputs and Life Cycle Inventory. Inputs for the petroleum LAS pathway shown in Figure 1 were provided by UOP. In the place of confidential UOP data, Table 1 summarizes petroleum LAS inputs from the ecoinvent Database for LAS, which are representative of but not identical to the actual inputs used for this study.[16] However, the UOP inputs are more representative of current technology than 2003 ecoinvent profiles. Life cycle inventory data corresponding to the inputs in Table 1 are from ecoinvent ecoprofiles, as shown in Table S1 of the Supporting Information. An average United States electricity grid mix was used for petroleum LAS production assuming a United States production location.

Table 1. Representative Inputs for 1 kg of LAS Production from Petroleum (from the ecoinvent Database)[a]

input	amount	unit
benzene	0.251	kg
paraffin	0.516	kg
sodium hydroxide	0.127	kg
sulfur	0.1	kg
catalyst	0.02	kg
heat	3.78	MJ
coproducts	see Figure 1	

[a]Confidential UOP inputs are within ±20% of the given data.

Inputs for the production of CO and PKO were taken from recently published studies from India and Malaysia, as these countries are major producers of coconut and palm products.[12−14,17] All input data were assembled into process stages within SimaPro 7.2 using ecoinvent ecoprofiles, as was also done for the petroleum pathway. Tables 2, 3, and 4 show the inputs for cultivation and harvesting, oil extraction, and conversion of the biorenewable oils to normal paraffins, respectively, the latter by means of UOP's catalytic deoxygenation technology.[18] Table 4 provides formerly published inputs that are representative of the confidential data used in our work for this stage. Life cycle inventory data corresponding to the inputs listed in Tables 2−4 are shown in Tables S2−S4 of the Supporting Information. N_2O emissions were estimated using the "tier 1" IPCC factor for direct and indirect mechanisms, 0.01325 kg N in N_2O/kg N in fertilizer.[18] The extraction of CO and PKO is done mechanically, requiring only the inputs of electricity and transportation (Table 3).[12,13] In Table 3, the inputs for oil extraction for coconut and palm kernel are unallocated, and because palm kernel is a small coproduct from the palm oil pathway, these extraction inputs for PKO are much larger than for CO. Electricity for the extraction of PKO and CO are accounted for by their respective country electricity grid mixes for Malaysia and India.[19]

Table 2. Inputs for the Cultivation and Harvesting of 1 kg of Renewable Feedstock (unallocated)

	coconut cultivation		palm cultivation	
input	amount	unit	amount	unit
ammonium sulfate, as N	0.0022	kg	0.0018	kg
urea, as N	0.0011	kg	0.0018	kg
single superphosphate, as P_2O_5	0.0021	kg	–	kg
potassium chloride, as K_2O	0.008	kg	–	kg
diesel	0.0022	kg	0.0037	kg
transport	0.0547	tkm	–	tkm
urea carbonyl group emission	0.0011	kg	0.0039	kg
N_2O emission	0.0033	kg	0.003 65	kg

Table 3. Inputs for the Extraction of 1 kg of Coconut and Palm Kernel Oils (unallocated)

input	coconut oil		palm kernel oil	
fruits	4.7	kg	43.13	kg
electricity	0.172	kWh	0.265	kWh
fuel oil	0.297	MJ	–	MJ
transport	0.134	tkm	0.317	tkm
coproducts	see Table S5 of the Supporting Information for complete list			

Table 4. Representative Inputs for the Production of 1 kg of Renewable Paraffins[18]

input	amount	unit
refined CO or PKO	2.033	kg
hydrogen	0.058	kg
hydrocarbon solvents	0.006	kg
electricity	0.092	kWh
fuel gas	0.0116	kg
boiler feed water	0.66	kg
cooling water	6.401	kg
coproducts	see Table S5 of the Supporting Information for complete list	

Various transportation steps for the movement of intermediate products have been handled in this study in the following manner. The palm oil extraction mill is assumed to be on the palm plantation, and thus, no transport is required to move biomass, while transport of the kernels to the palm kernel extraction mill is accounted for by truck transport over 86.8 miles in the extraction stage.[12,13] Coconuts are transported 34 miles by truck to the CO mill. Paraffin and LAS production from the extracted oil are assumed to be carried out in the United States, which has been accounted for by truck transport of the oil a distance of 83.3 miles to a port for CO, and with the PKO mill assumed to be at the port, 9000 miles of transoceanic transport to the United States and truck transport of oil from the United States port to the LAS production facility of 50 miles has been assumed.

LAS Use. We do not include in the scope of the LCA formulation of LAS the packaging and use in the home because they are common to both the petroleum and renewable LAS pathways.

End of Life Emissions. After end use, LAS typically becomes a contaminant in municipal wastewater. EoL emissions for this LCA encompass the LAS-specific emissions generated from the wastewater treatment (WWT) processes and aerobic degradation after application of the resulting sludge to land. We do not include in this LCA the inputs to WWT because the allocation

Table 5. GHG Emissions Generated from LAS with PKO, CO, and Petroleum-Based Paraffins Assuming Mass Allocation

paraffin source	cradle-to-gate emissions (kg CO_2 eq/kg LAS)	EoL emissions (kg CO_2 eq/kg LAS)	sequestration (kg CO_2 eq/kg LAS)	cradle-to-grave emissions (kg CO_2 eq/kg LAS)	reduction in life cycle emissions compared to petroleum LAS (%)
PKO	1.15	0.81	0.032	1.93	46.2
CO	1.11	0.81	0.032	1.89	47.3
petroleum	1.22	2.36	0.00	3.58	–

of these inputs to LAS versus other components of wastewater are the same for both petroleum and renewable LAS. One study covering nine sewage treatment facilities and three different design systems indicates that approximately 98% of LAS is mineralized to CO_2 during WWT and application of sludge to land, while 2% is sequestered in the soil after being applied to fields.[8] Consistent with IPCC, and with EPA and EU RED legislation, biogenic carbon is assumed to be carbon neutral, and therefore, the EoL emissions for renewable LAS do not count toward the carbon footprint from the LAS degradation, only from the fossil-derived benzene ring portion of the molecule. In contrast, the petroleum surfactant's EoL emissions must include all carbon dioxide emitted due to degradation. For sequestration in soil, only the biogenic carbon is counted and is assigned a negative emission but not so the carbon atoms from fossil sources, which were originally sequestered in the earth before processing. A sample calculation of EoL emissions for petroleum-derived LAS is shown in the Supporting Information.

Allocation. Many of the process stages in the production of LAS generate coproducts (Figure 1). The coproducts from the preparation of CO are coconut shell and meal.[14] PKO production gives empty palm fruit bunches (EFB), palm meal, palm oil, and palm kernel shells as coproducts.[12,13] Coproducts for paraffins derived from fossil kerosene include heavy, medium, and light kerosene. Renewable paraffins production from CO and PKO include coproducts such as renewable fuel gas, LPG, naphtha, and diesel (Figure 1 and Table S5, Supporting Information). The primary coproducts from the production of LAS from each paraffin source include fuel gas, liquid hydrocarbons, and other aromatic hydrocarbons. Inventory elements from each stage, and all prior stages, are shared between the main product and the coproducts through an allocation method. The base case of our study uses mass allocation to distribute the inventory between the individual products of each stage. An energy allocation scenario was also developed with the allocation factor modified by multiplying the mass of each product and coproduct by its respective lower heating value (LHV) in MJ/kg.[10,20−22] A sample calculation for mass allocation is shown in the Supporting Information, and the LHVs used in the energy allocation are presented in Table S5 of the Supporting Information along with a listing of all allocation factors in Table S6. A market value allocation scenario based on the economic value of the system products and coproducts for the base case was also performed, the data for which is presented in Table S13 of the Supporting Information.

Impact Assessment. The impact category chosen for this study is greenhouse gas (GHG) emissions. The IPCC 2007 GWP 100a method in SimaPro 7.2 was used to determine GHG emissions associated with LAS production. Global warming potentials were applied to place each major chemical's emissions on a basis of kg CO_2 equivalents (eq)/kg LAS. For instance, methane and N_2O emissions are placed on a CO_2 equivalent basis using global warming potentials of 25 and 300, respectively.

Results and Discussions for LAS and dLUC. The greenhouse gas emissions of LAS for cradle-to-gate, EoL, and cradle-to-grave system boundaries for each paraffin source are included in Table 5 assuming mass allocation. This table also shows percent reduction of GHG emissions compared to petroleum LAS. Table S7 of the Supporting Information includes the same GHG results for energy allocation. Table S8 of the Supporting Information presents the contributions to GHG emissions from the various transportation steps. The cradle-to-gate GHG emissions, not including EoL, for production of LAS are within 10% for all paraffin sources, with the GHG emissions from the renewable sources being lower than production from petroleum. Emissions of greenhouse gases from EoL processing (wastewater treatment) for renewable LAS are approximately a third of the EoL emissions from petroleum LAS. Net GHG emissions for PKO and CO LAS with mass allocation are 1.93 and 1.89 kg CO_2 eq/kg LAS, respectively, which are savings of greenhouse gas emissions of 46.2% and 47.3% compared to petroleum LAS (3.58 kg CO_2 eq/kg LAS), respectively. It can be concluded that the savings of GHG emissions of renewable LAS are due almost entirely to the difference in how EoL emissions are counted for the renewable and fossil carbons in LAS.

When energy allocation is used rather than mass allocation, more of the emissions from the pre-LAB production stages are attributed to renewable LAS resulting in ∼1.1% to ∼1.5% percentage points less savings for coconut and palm kernel derived LAS, respectively. This small change is due to the larger lower heating values (LHVs) of CO and PKO compared to the other coproducts produced during oil extraction. For the petroleum LAS pathway, GHG emissions for mass and energy allocation show only a 2% difference because all coproducts and products have similar LHVs. The effect of allocation method makes a 4% maximum difference on the LAS emission results between mass and energy allocation.

Figure 2 shows the stage-by-stage contribution to the overall life cycle GHG emissions for petroleum-, CO-, and PKO-based LAS with neutralization assuming mass allocation. Figure S1 of the Supporting Information shows the same set of stage GHG results assuming energy allocation. While the paraffin production, LAB production, and Neutralization stage emissions shown in Figure 2 are significant to the total GHG emissions, the EoL emissions are the largest differentiator between the emissions from the renewable and petroleum LAS pathways. Both CO- and PKO-based LAS reduce the greenhouse gas emissions compared to petroleum LAS by greater than 40% because of the reduced emissions at EoL. These stage-by-stage GHG results indicate that a high certainty in EoL emissions of LAS is critical to affirm the GHG emission savings of renewable LAS. Effects of variations in EoL fate of LAS are investigated in a scenario analysis.

Land use change can have a significant impact on the emissions of biorenewable feedstocks as carbon stocks in plant

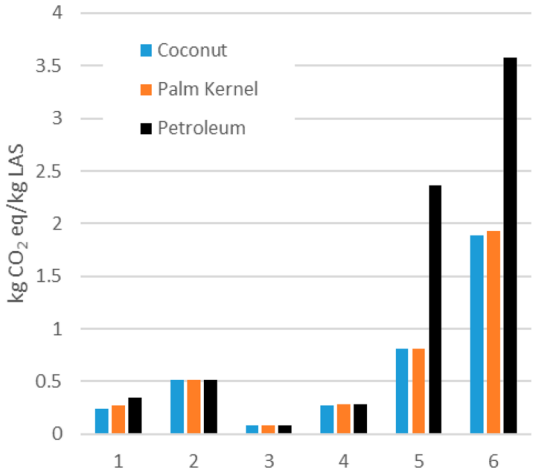

Figure 2. Stage-wise greenhouse gas contributions for the production of LAS from PKO-, coconut oil-, and petroleum-based paraffin sources assuming mass allocation: (1) paraffin production, (2) LAB production, (3) sulfonation, (4) neutralization, (5) EoL, and (6) total.

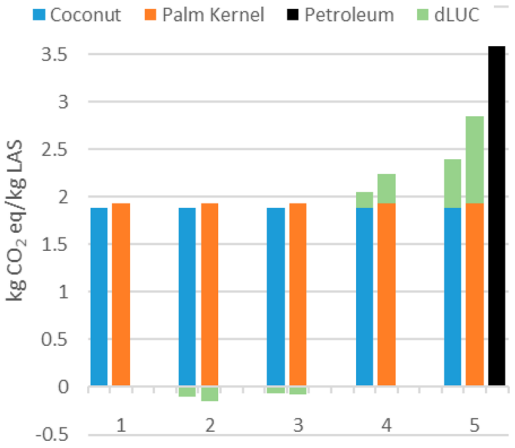

Figure 3. dLUC contributions to life cycle emission by land type assuming mass allocation: (1) base case (no dLUC), (2) grassland, (3) shrubland, (4) secondary Forest, and (5) primary forest.

matter, and soil can vary significantly from the original land cover compared to the renewable feedstock crop.[23] This change of carbon stocks can result in increased sequestration of CO_2 in soil (counted as a CO_2 emissions credit), reduced sequestration (counted as emission of CO_2), or no net change in carbon stocks. Because our study assumes expansion of plantations using noncrop lands, our study does not account for indirect land use change CO_2 emissions and therefore focuses on direct land use change (dLUC) only. As we assume that sustainable practices will be followed, peatland is not considered a viable option for conversion to plantations and is thus not included in the dLUC calculations. The dLUC emissions for this study were calculated according to the 2BSvs and ISCC 205 GHG methodologies.[24,25] In each methodology the mature carbon stock of the crop of interest is subtracted from the carbon stock of the previous land occupation or reference carbon stock. This carbon stock difference is then converted to CO_2 equivalents, divided by the period of analysis, and divided again by the productivity of the crop. The dLUC equation used in the current study is shown in the Supporting Information.

Carbon stock numbers were obtained from literature sources with an average value being used for both crops planted in four land use categories: primary forest, secondary forest, shrubland, and grassland.[22,26−28] Crop productivity is an average of literature values for oil palm and obtained from Sreejith et al. for coconut[14] (see the Supporting Information for details, specifically Table S9). Calculated dLUC GHG impacts for mass allocation shown in Figure 3 indicate that using primary forest land for plantations results in a significant increase in emissions compared to results in Table 5, while using secondary forest land generates at most a 26% emissions increase from the base case. Either shrubland or grassland results in a maximum sequestration of carbon for dLUC of 12%. Table S10 of the Supporting Information shows the values displayed in Figure 3. These results show that cultivation for production of CO and PKO should avoid primary forest lands.

In the calculation of dLUC emissions, energy allocation assigns more of the dLUC emissions toward the CO and PKO products in the pre-LAB production stages resulting in a larger increase to the life cycle contribution. Figure S2 and Table S11 of the Supporting Information present the dLUC GHG emissions under energy allocation. The dLUC results were generated using average values for C stocks and plantation yields, and significant variability in dLUC results can be expected from location to location.

Scenario Analyses Variation in EoL Emissions, Alternate Electricity Mixes from United States Regions, Market Value Allocation, and Comparison to FAS. Four scenarios were considered: the effect of variations in the EoL emissions on the total emissions of LAS to account for uncertainty, the effect of alternate NERC region electricity mixes, allocation by market value, and a comparison of renewable LAS to similar surfactants such as fatty alcohol sulfates (FAS) produced from the same feedstocks.

The sensitivity of the greenhouse gas emissions to the fate of LAS at the EoL was examined for the mass allocation base case. According to Berna et al., between 90% and 99.5% of the spent LAS is mineralized to CO_2, with the most likely value of 98%, which was used in the base case.[8] At 90% of carbon mineralized to CO_2, palm and coconut LAS increase in savings from 46.2% and 47.3% (Table 5) to 48.8% and 50%, respectively. Conversely, when 99.5% of carbon is mineralized to CO_2, palm and coconut LAS decrease in savings to 45.7% and 46.8%, respectively. Table S12 of the Supporting Information shows the GHG emissions for the EoL scenarios. Over the range of variation studied here for EoL emissions, there is a maximum ±10% change in the renewable LAS GHG emissions, which translates into only ±2.7 percentage points of savings relative to petroleum LAS.

The choice of electricity mix can often have a significant effect on the life cycle emission results in biofuel or bioproduct LCA; thus, a scenario analysis was conducted on the PKO pathway (mass allocation) base case to determine how a different electricity mix may have changed the results of the study. The North American Electric Reliability Corporation (NERC) regions NEWE, ERCT, and SRMW were chosen to represent low, average, and high emissions relative to the United States eGrid average, respectively, which resulted in ±3.20% of the base case total emissions.[29] Sample calculations are shown in the Supporting Information, and data are shown in Table S14. In this study, the electricity mix has only a small effect on emission results.

Market value allocation (MVA) distributes the burdens between coproducts based on their economic value. Prices were

found for the coproducts from the LAS production pathway and used to allocate GHG emissions; pricing data and allocation factors are shown in Table S13 of the Supporting Information. This resulted in an increase of ~9.3% kg CO_2 eq emissions/kg LAS from renewable LAS and a corresponding decrease in savings to 41.1% (5 percentage points).

FAS is a surfactant similar to LAS that can be produced from the same renewable feedstocks. It is an alkane chain with an average chain length of 13 carbons with a sulfate group at one end. SimaPro 7.2 includes ecoinvent profiles for both biorenewable- and petroleum-based FAS with inventories developed based on mass allocation for coproducts, which makes these FAS profiles a good comparator for the LAS in this study. Table S15 of the Supporting Information shows the GHG emissions of FAS derived from CO, PKO, and petroleum. EoL emissions assumptions for FAS are the same as the base case for LAS, 98% mineralization of carbon to CO_2. Figure 4 presents the total GHG emissions from FAS alongside

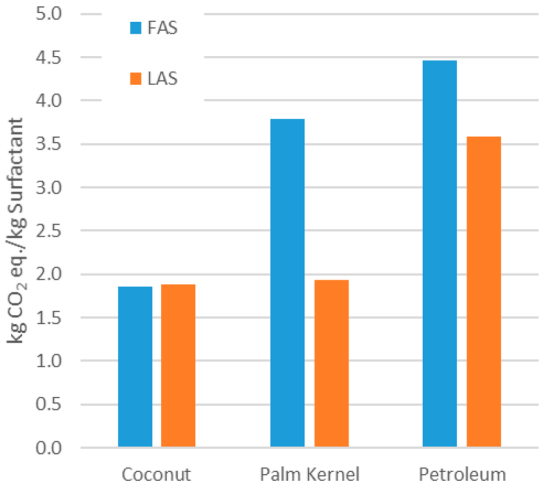

Figure 4. GHG emissions for FAS and LAS with mass allocation.

those for LAS for mass allocation. The life cycle GHG emissions for LAS are approximately equal to FAS for CO-derived product compared to FAS and are 51% lower than PKO-based FAS. FAS from PKO and CO exhibit lower GHG emissions compared to fossil FAS. It must be noted however that any comparisons between renewable or petroleum LAS to FAS are very preliminary and subject to large uncertainty due to differences in quality of the inventory data in terms of geographic, time, and technology relevance.

The global chemicals industry is seeking ways to move toward sustainable production in ways that cost effectively satisfy consumer demands and with environmental and societal benefits. Transitioning from fossil-based to renewable-based production systems has been promoted as one strategy to achieve sustainability. This study demonstrates that when renewable molecules substitute for fossil molecules in LAS, large savings of GHG emissions are achieved. CO_2 emissions during end of life processes were shown to be the key differentiator among the LAS alternatives.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Sample calculations, expanded results tables, tables of data used in allocation and dLUC calculations with references, and detailed energy allocation results. These also include data for the electricity mix and market value allocation scenarios. This material is available free of charge via the Internet at http://pubs.acs.org.

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. D. P. Fogliatti and S. A. Kemppainen contributed equally.

Notes

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