Advanced Review

The liquid carbon challenge: evolving views on transportation fuels and climate

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Carbon-based liquid fuels are highly valued for transportation; they are the world's largest form of commercial energy and second largest source of anthropogenic carbon dioxide (CO_2) emissions. Strategies to address their CO_2 emissions have been shaped by fuel cycle analysis (FCA), a version of lifecycle assessment that examines fuel products and their supply chains. FCA studies have diverse findings and large uncertainties. Disagreements are particularly sharp for biofuels, which are seen as key replacements for petroleum fuels. A critical reading of the evolving literature reveals problems of model structure, including system boundary misspecification, flawed carbon cycle representation, and use of a static framework to analyze dynamic systems. New analytic paradigms are needed for liquid fuels, given their tradability, the realities of the carbon cycle, and the implausibility of capturing carbon from mobile sources. Logical decomposition of options shows that, beyond measures to limit fuel demand, CO_2 emissions from liquid fuels must be balanced by increasing the rate of net carbon fixation. Further analysis and discussion are needed of carbon accounting methods, energy research priorities, ways to link CO_2 removal options to fuel-related mitigation efforts, and the transportation elements of climate policy. © 2014 John Wiley & Sons, Ltd.

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INTRODUCTION

Carbon-based liquid fuels are the world's largest source of commercial energy and expected to stay so for at least the next two decades.[1] Petroleum (conventional and unconventional crude oils as well as natural gas liquids) is projected to remain by far the main source of liquid fuels. Liquid hydrocarbons are ideal mobile energy carriers; other liquid organic compounds such as alcohols come close. Although other uses of liquid fuels have been reduced over time, widespread replacement of liquids in transportation remains difficult in spite of years of effort to develop options using electricity, hydrogen, or natural gas. Non-carbon-based energetic liquids such as ammonia or hydrazine exist, but have very limited use (e.g., rocket propulsion). The energy density and fluidity that offer such convenience for motor vehicles, boats, and aircraft also make carbon-based liquid fuels, and their primary feedstocks, crude oils, easy to trade globally. They are the focus of this article and inspire the 'liquid carbon' shorthand of the title.

Biofuels (here referring to liquid energy carriers derived from biomass, as opposed to solid or gaseous forms of bioenergy) are widely seen as crucial for replacing fossil-derived liquids in a climate-constrained world.[2] They have strong support through the influence of agricultural interests. Biofuels also enjoy political appeal under the banner of energy security and the belief that, unlike fossil fuels, biomass feedstocks are a renewable resource. Traditional industrial biofuels, such as ethanol from grains and sugarcane or biodiesel from oilseeds, have seen rapidly rising production over the past decade. Although their use may continue to grow under current policies, biofuels still account for less than 2% of world liquid fuel consumption.[3]

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As a source of carbon dioxide (CO_2) emissions, liquid fuels are second to coal and accounted for an estimated 3.2 PgC/year (33%) of the total anthropogenic CO_2 emissions of 9.7 PgC/year as of 2012.[4] By convention, such greenhouse gas (GHG) inventory tallies exclude CO_2 emissions from biofuel combustion.[5] The assumption is that, because the biomass feedstocks absorb CO_2 from the air, biofuels are inherently 'carbon neutral' and so replacing fossil carbon in a fuel with biogenic carbon yields a one-for-one reduction in CO_2 emissions (other than production-related emissions). However, the CO_2 directly emitted from liquid fuel combustion varies very little per unit of delivered energy. Therefore, biofuels do not actually reduce CO_2 emissions downstream in end-use sectors (such as transportation) where fuels are used. Any mitigation effect occurs elsewhere (upstream), in the agriculture and forestry sectors where feedstocks are grown.[6]

The benefits of biofuels have been disputed for many years. Even before GHG concerns were on the table, when energy security was the main driver, questions about biofuels' production energy requirements, the food versus fuel trade-off, the need for cellulosic processing technology, harvest-related price volatility, and economic viability were debated.[7] Many such discussions involved net energy analysis, which examines fossil energy use throughout a fuel's supply chain relative to the energy value of the fuel[8,9] and analyzes different fuels 'on an equal work provided basis'.[10]

Net energy analysis evolved into 'well-to-wheel' models for fuel-related lifecycle analysis (LCA), or fuel cycle analysis (FCA) as it is termed.[10–12] That technique has been applied to evaluate the energy and GHG emission impacts of a wide variety of existing and proposed fuels, including fossil options (coal-to-liquids, gas-to-liquids, unconventional petroleum) as well as electricity, gaseous fuels, and biofuels from a range of feedstocks. LCA-based GHG evaluations of many products and activities (not just fuels) also came into vogue under the rubric of 'carbon footprinting'.[13] A now almost universal recommendation (often simply a presumption) is that fuels should be compared according to lifecycle impact.[14–20] An extensive literature exists on the subject, including summaries with representative results,[17,20,21] critical reviews,[22–25] and government-sanctioned findings.[26–28] The latter are generated in service of public policies designed to promote biofuels, including US Renewable Fuel Standard (RFS),[26] the California Low-Carbon Fuel Standard (LCFS),[27] and the EU Renewable Energy Directive (RED).[28,29]

In spite of the apparent maturity of FCA methods, views on the subject, particularly on the role of biofuels as a 'low-carbon' replacement for petroleum fuels, seem no closer to consensus today than they were a generation ago. Indeed, since researchers[30,31] quantified the carbon stock impacts of land-use change tied to globally growing demand for food, feed, fiber, and fuel, the debates have become even more fraught. Analysts have noted FCA's methodological challenges[22,32–34] and there were even earlier critiques of the net energy analysis method from which FCA was derived.[35] Some researchers involved in developing and applying FCA recently highlighted the method's large uncertainties and the likelihood of misleading results.[36–38] Nevertheless, many researchers view FCA as a method of choice and advocate efforts to further the technique.[19]

The persistent disputes over FCA results suggest a need to re-examine basic assumptions and scrutinize the method more deeply than has been done in previously published reviews of data inputs, parameter choices, and model selection. FCA is not the only approach for evaluating GHG impacts of fuel use; it can be compared to other methods of analysis and to integrated assessment modeling (IAM). Such comparisons reveal that a critical issue is not just that of how FCA treats land-use change, but rather about how land itself is treated when defining the system boundaries for analysis. Before turning to that discussion, this article briefly describes the scale of fuel-related CO_2 emissions. It then examines FCA and other analytic methods, leading to a discussion that clarifies the liquid carbon challenge and suggests directions for future research.

CO_2 EMISSIONS FROM FUEL USE

Long-term tracking of national and international CO_2 emissions from fossil fuel use, cement making, and land-use change is carried out by the Carbon Dioxide Information and Analysis Center (CDIAC)[39] and analyzed through the Global Carbon Project (GCP).[40] Figure 1 shows global trends in anthropogenic CO_2 emissions since 1990. Emissions from liquid fuels rose slowly over the period, recently reaching just over 3 PgC/year (simply labeled here as 'oil' and referring to all fossil-based liquid fuels including those derived from bitumen and natural gas liquids). Coal overtook oil as the leading source of CO_2 emissions in 2005. On a global basis, the direct CO_2 emissions from biofuel combustion are balanced by CO_2 uptake during feedstock growth and therefore do not show up in Figure 1. Any release of carbon stocks that might be induced by rising production of biofuel feedstocks falls

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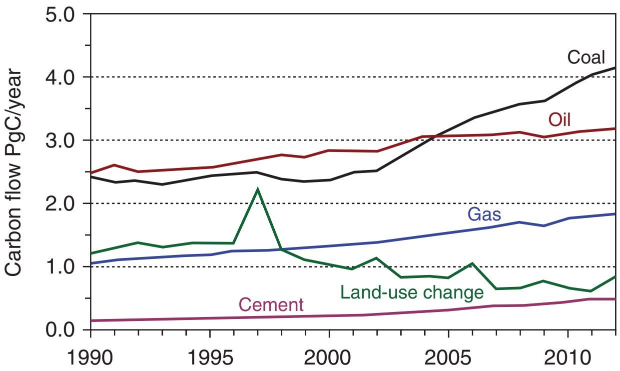

FIGURE 1 | Global anthropogenic CO_2 emissions, 1990–2012. Here coal refers to solid fossil fuels; oil refers to liquid fuels including those derived from conventional and unconventional petroleum as well as natural gas liquids; gas refers to natural gas and other gaseous fossil fuels plus gas flaring. Source: CDIAC.[39]

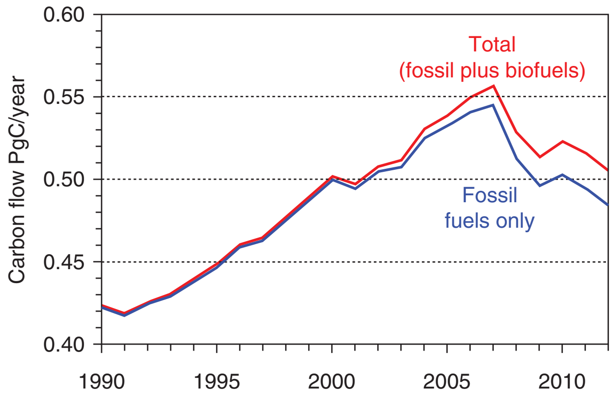

FIGURE 2 | Direct CO_2 emissions from US liquid transportation fuel use, 1990–2012. Source: EIA Monthly Energy Review.[43]

within the land-use change category shown on the chart.

Although a biofuel lifecycle is inherently carbon neutral when narrowly defined, it may not be so when land-use change is considered (even aside from production-related emissions, which are yet another matter). Moreover, CO_2 emissions from biofuel use may not be balanced by CO_2 uptake within the confines of a national-scale GHG inventory and certainly are not balanced within an energy use sector such as transportation.[41] The reason has to do with questionable net uptake at any subglobal level and the fact that CO_2 uptake occurs in land-use sectors at locations remote from motor vehicles and other points of fuel end-use.[6] This accounting disparity is problematic, given that climate policies are administered at national levels and with different (or absent) levels of accountability across sectors and across international borders.[42] Physically within energy end-use sectors, biofuel combustion is a source of CO_2 that directly enters the air, adding carbon to the atmosphere in a manner indistinguishable from the CO_2 emitted by fossil fuel combustion.

This perspective is shown in Figure 2, which plots CO_2 emissions from liquid fuel consumption in the US transportation sector[43] (on a carbon mass basis as in Figure 1). The lower (blue) curve shows direct (end-use) CO_2 emissions from fossil-based fuels (largely gasoline, diesel, and jet fuel). Only such fossil-based CO_2 is included in official GHG emissions inventories and reports on progress in GHG control. The upper (red) curve adds in end-use CO_2 emissions from biofuels (ethanol and biodiesel), which reached 4.3% of the total 0.51 PgC/year as of 2012. The recent increase in biofuels is largely accounted for by corn-based ethanol. Counting only the fossil portion per GHG inventory conventions, liquid transportation fuels accounted for 27% of the total US GHG emissions, which were 1.86 PgCeq/year (all gases, carbon mass basis) in 2010.[44]

Complex FCA methods have been used to calculate CO_2 emission reductions from substituting biofuels for fossil fuels; such modeling underpins claims of climate benefits for the RFS and LCFS. For example, the US Environmental Protection Agency (EPA) projects an annualized emissions reduction of 138 million metric tons (CO_2-equivalent; equal to 0.038 PgC) for the RFS if it is fully implemented at 36 billion gallons of renewable fuel in 2022.[26] The analyses behind such estimates account for land-use change, but the uncertainties are very large.[45] The net effect on global GHG emissions can be ambiguous, with results greatly dependent on the path of implementation as well as modeling assumptions regarding agricultural trade, crop management, yields, and land-use responses.[46,47] Some studies conclude that it is impossible to quantify a claim of GHG reduction with any confidence.[48,49] Moreover, the uncertainties involve not only issues of inadequate or conflicting data, which might eventually be resolved through empirical work, but also system boundary definitions and other modeling conventions. It is to those issues that this discussion turns next.

OVERVIEW OF ANALYTIC METHODS

Both research priorities and public policies for mitigating GHG emissions from transportation fuels are influenced by the methods used for analysis. Four main approaches have been used:

(a) FCA, a form of LCA applied to transportation fuels and as implemented in GREET and similar

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models[11,12,20,24,50]; FCA is now used for compliance purposes in policies such as the US RFS, California LCFS, and EU RED.

(b) Terrestrial resource analysis (TRA) methods based on ecology and forest management research, as seen in the standard methodology described by Schlamadinger et al.[51]

(c) 'Kyoto accounting', the method for tallying GHG sources and sinks formalized by the IPCC for reporting of national GHG emissions inventories and as used in the design of cap-and-trade programs and other aspects of international climate policy.[5]

(d) IAM methods, which examine bioenergy systems in the broader context of combined climatic and economic system modeling at the global level.[21,52]

These approaches were developed for different purposes and to a notable degree evolved within different disciplinary traditions.

Fuel Cycle Analysis

Fuel-oriented LCA has been very influential in shaping both scientific thinking and public policy regarding GHG mitigation strategies for transportation fuels, and so it is a prime focus of this review. LCA was originally developed to assess environmental impacts of many dimensions, not generally focusing on fuels or climate impacts.[53] The technique has matured over the years, with an International Standards Organization (ISO) series that lays out principles, guidelines, and interpretative considerations in great detail.[54]

Fuel-oriented versions of LCA developed in the late 1980s and early 1990s. FCA grew out of neither the general LCA nor the geoscience traditions, but rather out of applied physics and energy engineering studies spurred by the 1970s oil crisis. The motivation was finding ways to minimize reliance on fossil resources, especially petroleum, leading to the use of net energy analysis as seen in early evaluations of alternative fuels.[8,55] DeLuchi (now Delucchi) pioneered FCA at the University of California, Davis, and in a major study for Argonne National Laboratory (ANL) sponsored by the US Department of Energy.[11] That work presaged LEM,[56] GREET,[50] and similar models[24] now in widespread use. Rooted in net energy analysis, these methods are at heart 'bottom-up' engineering models that apply emissions factors (e.g., in gCO_2e/MJ) to calculate GHGs and other impacts for each step in a supply chain.[11,12,57]

Methodological Orientation

LCA can be classified as *attributional* (ALCA) or *consequential* (CLCA). As the name suggests, ACLA attributes impacts to a particular product system. It restricts system boundaries to 'flows physically connected to the product under study' and is static in that system dynamics are not represented, and it does not 'account for price variations, changes in demand or technological improvements'.[58] (To clarify this point, LCA does not model the process of technology change even though it is often used to evaluate prospectively assumed technology developments.) In contrast, CLCA examines the consequences of different systemic choices, modeling dynamic and behavioral effects using a 'system-wide approach where system boundaries are expanded [to] evaluate all of the changes in a system as a consequence of a decision'.[58]

FCA is fundamentally attributional but in practice often includes partly consequential elements for addressing coproducts. FCA is interpreted as an ALCA, using a fuel product system as the basis for defining system boundaries and attributing a lifecycle GHG emission impact, or 'carbon intensity' (CI), to different fuels.[27,50,59] Fuel product systems are called *pathways* and represent technology and processing linkages among feedstocks, fuel products, and the associated inputs and coproducts.[57] FCA uses empirical factors for process technology and other production impacts such as non-energy-related emissions (CH_4 from various sources, N_2O from soil, etc.). Distinct pathways (crude oil to gasoline, corn to ethanol, soy to biodiesel, or multiple resources to blended fuels or electricity) are modeled separately and then compared.[59,60] Such FCA results are widely reported in the literature; e.g., see Figure 2.10 of the IPCC Special Report[21] for typical results (excluding indirect land-use change effects, as discussed later).

The system boundaries used to treat key carbon flows in FCA are illustrated in Figure 3. This diagram shows only the main processes affecting CO_2 emissions; FCA models address many other effects and a complete schematic would be more complex. Nevertheless, it highlights critical features of the FCA framework, which is seen as providing a 'complete side-by-side LCA of biofuel production relative to fossil fuel production'.[25] The petroleum-based fuel pathway (1) addresses all processes and associated emissions starting with the extraction of crude oil through refining and on to fuel combustion (some steps, such as oil shipping, are omitted here for simplicity). The biofuel pathway (2) also addresses all emissions starting with farming (including direct land-use change) and then processing through combustion.

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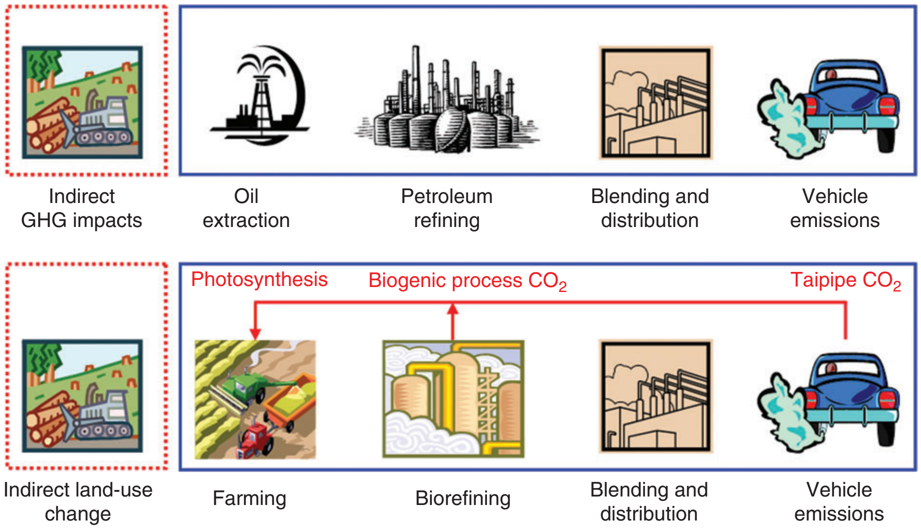

FIGURE 3 | System boundaries as commonly defined for fuel cycle analysis. (a) Petroleum fuel and (b) biofuel.

For biofuels, FCA assumes that biogenic CO_2 emissions, both from end-use combustion and during processing (such as burning crop residues for energy), are fully balanced by CO_2 uptake during feedstock growth. Such automatic crediting reflects the view that CO_2 emissions from biofuel combustion need not be counted 'because they are not net emissions to the atmosphere'.[61] This convention is now ensconced in public policy, e.g., as stated by EPA for the RFS, 'CO_2 emissions from biomass-based fuel combustion are not included in their lifecycle emissions results'.[62] As seen in Figure 3(b), the carbon neutrality assumption is arithmetically correct within a biofuel lifecycle. It is also true globally if *all* biomass used in the world is the subject and terrestrial carbon stock impacts due to land-use change are accounted for separately. But it is not necessarily true for any particular fuel product system or for biofuel use at a national or other subglobal level.

Treatment of Land Use

All commercially proven biofuel feedstocks are land intensive and therefore their production creates a demand for arable land. Land that supports high yields by virtue of ample sunshine, moisture, and other features conducive to agriculture and forestry is a finite resource. Biofuels compete with other agri-forestry demands and expanding their production puts upward pressure on commodity prices.[63] That in turn motivates new land conversion, including market-induced impacts in frontiers remote from feedstock production.[31,64–66] Although the causal chain for any such indirect land-use change (ILUC) is complex, most new land conversion impacts tropical forests.[67] Whether direct or indirect, the resulting deforestation releases a great deal of carbon, marginally increasing the land-use change emissions trend shown in Figure 1 but representing a near-term impact that is highly uncertain but could be many times larger than the CO_2 emissions from fuel combustion per unit of energy.[31,49,66] The result is *carbon debt*,[30] reflecting the number of years it takes for the annual emissions reduction imputed to biofuels to compensate for, i.e., 'pay back', the CO_2 released during land-use change due to an expansion of production.

As shown by the dashed border in Figure 3, ILUC falls outside the FCA system boundary as conventionally defined. Quantifying ILUC requires consequential analysis at a global scale, involving commodity trade models coupled to land-use models.[27,62,68–70] This entails scenario analyses that span forward over an assumed time horizon (e.g., 20 or 30 years) and the results are reduced to a single number, an ILUC factor in gCO_2/MJ, that is added to the results from the underlying LCA. These variable projections are now the dominant uncertainties confronting FCA.[45–48]

Although summing attributional plus partially consequential modeling results yields a metric that is arguably incoherent as well as scientifically indeterminate, such an approach is used to generate the CI values for regulating fuels under the LCFS.[27] EPA's RFS modeling provides an even more complex consequential analysis of not only ILUC but also many other substitution and price effects for coupled energy and agricultural markets, and the agency initially reported its results as scenarios over an assumed lifecycle.[62] Nevertheless, the RFS approach remains attributional in orientation and reduces the results to CI values (gCO_2e/MJ) similar to those used in the LCFS and throughout the FCA literature.[26]

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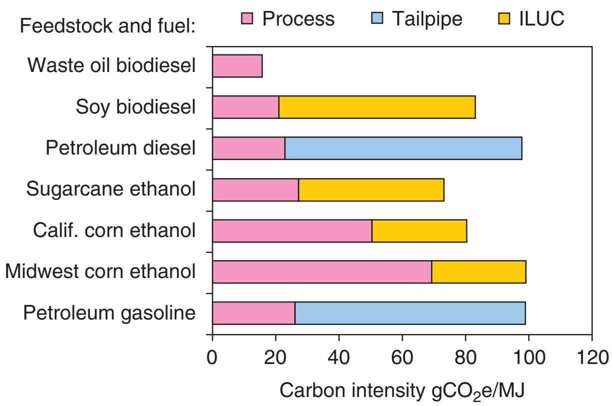

FIGURE 4 | Illustrative fuel cycle analysis results for some existing fuels. ILUC = indirect land-use change. Source: CARB LCFS lookup tables, December 2012.[71]

Illustrative FCA Results

Figure 4 shows illustrative CI values for several fuels; the purpose here is not to scrutinize these numbers *per se* but rather simply to show how such results are commonly reported. The leftmost (pink) portion of each bar gives the upstream GHG emissions directly tied to a fuel's supply chain, including feedstock production and processing as well as emissions credits for coproducts. Fuel combustion ('tailpipe', in blue) emissions are shown only for the fossil fuel products; for biofuels, they are zero under the carbon neutrality assumption.

Traditional FCA studies addressed only the first two portions of each bar (pink plus blue), and commonly found that biofuels had a lower lifecycle GHG impact than petroleum fuels, in some cases by a large margin. For the examples shown,[71] ethanol from Midwestern corn has a direct CI 30% lower than that of gasoline and ethanol from Brazilian sugarcane is 72% lower. Biodiesel from US soybeans has a CI 78% lower than petroleum diesel according to these estimates, which are consistent with prior research showing its merits due to lower agricultural and processing energy inputs.[72] The CI of biodiesel derived from waste oils is lower still.

As FCA was introduced in the early 1990s, numerous studies have been published and until 2008 nearly all the work assessed only impacts having physical links to a fuel's supply chain (i.e., excluding ILUC). The literature covers myriad feedstock and fuel combinations, including both current and prospective fuel pathways. Reviews of these studies highlight the variability and uncertainties related to differing databases, process efficiencies and emissions factors, parameter choices, system boundaries, coproduct allocation, and other methodological details.[21–24,73] Although there was some dissent,[74] the FCA literature generally reinforces the view that, given sufficiently efficient production processes, even traditional crop-based biofuels have lower GHG impacts than fossil-derived fuels.[2,9,14,57,59]

The incorporation of ILUC, an emissions leakage that undermines the CO_2 reductions otherwise attributed through FCA,[75] greatly changes the picture. The impact is shown by the rightmost (yellow) portions of the bars in Figure 4. CO_2 emissions from tropical deforestation, which is the main frontier of land conversion, entail a very large initial release followed by several years of soil carbon release and then a smaller ongoing amount of foregone CO_2 uptake (see Figure VI.B.5-1 of EPA's discussion).[62] For US corn ethanol, a notable early ILUC impact estimate was 104 gCO_2/MJ when averaged over a 30-year horizon.[31] Subsequent analysis[76] using different data and modeling that included countervailing effects computed a corn ethanol ILUC impact of 27 gCO_2/MJ, similar to the values shown in Figure 4. Midwestern corn ethanol then appears no better than petroleum gasoline; the benefit of sugarcane ethanol drops from 72 to 26% and the benefit of soy diesel relative to petroleum diesel falls from 78 to 15%; only waste-based fuel is unaffected.

Uncertainty Challenges

The uncertainties in ILUC projections are very large. Plevin et al.[45] found a range of 21–142 gCO_2/MJ (95% central interval) through simulations of the key factors influencing ILUC for US corn ethanol. Uncertainties as large as those entailed in such modeling imply that FCA cannot determine with confidence the sign (positive or negative) of the change in emissions for many real-world biofuels relative to fossil fuels.

A more profound concern surrounds FCA because it involves modeling effects that span many years into the future. Therefore, even if current data limitations could be resolved, FCA results cannot be empirically verified.[6] One parameter choice, the assumed length of a lifecycle, underscores the conundrum. The examples in Figure 4 adopt a 30-year time frame over which ILUC and other effects are averaged, as assumed for the RFS and LCFS. Some European analyses have used a 20-year horizon[66] and averaging over this shorter lifecycle yields ILUC factors 50% higher than when assuming a 30-year horizon, illustrating how a single parameter choice hugely affects the results. Although biofuels face the greatest trouble, analytic challenges also confront FCA characterizations of electricity and other fuels that policies such as the LCFS attempt to address.[77] FCA has thus evolved from its original attributional—and now *de*

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jure—objective of comparing individual 'fuels' to a tool that *de facto* compares alternative future worlds envisioned for complex and globally coupled fuel product systems, with all the irreducible uncertainties that that entails.

Terrestrial Resource Analysis

During the same early 1990s time frame that FCA evolved, a parallel body of work developed for analyzing how to best use forests and other terrestrial resources for mitigating climate change while addressing demands for traditional agricultural and forest products.[51,78,79] TRA grew out of a forest management tradition and, unlike lifecycle methods, is anchored in the ecology of the carbon cycle. TRA is designed to handle carbon stocks and flows, counting both sources and sinks while evaluating effects of changing land use and trade-offs between harvesting and letting forests continue to grow.[79]

TRA is a bottom-up approach that starts with natural resources (such as land and fossil fuel reserves) as the basis for the system to be analyzed, including production and use of biofuels, fossil fuels, other products, and their associated inputs. It treats biomass resource pathways and fossil resource pathways as subsystems within a unified system. As stated by Schlamadinger et al.,[51] 'the full system consists of a bioenergy fuel chain … and a reference fossil fuel chain …' That article outlines the considerations, including care in specifying system boundaries, that are critical when comparing biofuels to fossil fuels. It does not appear that FCA methods adequately drew on this work, and in any event, the FCA models that came into widespread use do not adhere to the principles articulated for TRA.

The structural difference can be seen by comparing Figure 5, a simplified system boundary diagram for TRA, to Figure 3. Because it examines a unified system operating under different conditions (e.g., with or without biofuel production), rather than comparing two distinct systems (e.g., biofuel vs fossil fuel) as done in FCA, TRA is inherently consequential even in its most basic formulation. It defines the analysis so that the system operations to be compared 'produce the same level of goods and services'.[51] Otherwise put, TRA involves a dynamic (change-based) counterfactual analysis in contrast to a static comparison of distinct fuel pathways as performed using FCA. Other than this critical distinction, TRA is similar to FCA in that it accounts for the GHG impacts of all processing steps (including coproduct substitutions) for both biofuels and fossil fuels.

Notably in TRA, *land* is always within the system boundary, but it is omitted from the reference fossil fuel system in FCA. This is the key contrast between Figures 3 and 5: for the former (FCA), two different boundaries each enclose separate systems that produce different goods and services; for the latter (TRA), a single system boundary encloses two subsystems of a unified system that produces the same outputs. The omission of land from the reference system means that the systems compared in FCA do not produce the same goods from the land even though they may provide the same level of energy services (such as a unit of motor fuel energy). Critically, the CO_2 uptake that occurs on land when fuel is derived from the fossil resource is absent from FCA baseline. As for any prior biomass harvest that embodies this uptake, the realization that it can be displaced by biofuel production motivates analyses to address ILUC. Such consequential modeling incurs very large uncertainties but it does not correct for FCA's use of inconsistent system boundaries. TRA accounts for prior land use by design and, although it does not treat economically induced leakages such as ILUC, it avoids the gross error of a misspecified system boundary.

FCA's system boundary error obscures the fact that an effective gain in net ecosystem production (NEP) is required for a biofuel to reduce CO_2 emissions relative to a fossil fuel, as can be shown by examining an integrated biofuel and fossil fuel system.[6] If land is already removing CO_2 from the atmosphere for another purpose (food, feed, or sequestration), then shifting that land to biofuel production may not remove more CO_2 from the atmosphere. The effect of such shifts is directly represented in TRA, but mishandled in FCA because prior land use is omitted from the reference system boundary. Although displacement effects are considered in ILUC analyses, those adjustments may not compensate for the gross neglect of pre-existing terrestrial CO_2 uptake by FCA. Such cases are very significant because most existing biofuel production is based on crops (corn, soy, sugarcane, etc.) grown on land already in production and therefore already absorbing CO_2 from the atmosphere. Curiously, this lapse was not flagged in an article that called for greater engagement by ecologists even though the authors noted that LCA standards were not well-informed by ecological theory.[25]

TRA can model a lifecycle by integrating the combined biofuel and fossil fuel systems over time, yielding metrics notionally similar to CI values from FCA. Such an approach is used to develop 'carbon neutrality' (CN) factors as proposed in some European discussions.[80,81] Being a dynamic model, TRA parametizes time; it also handles carbon debt from direct (though not indirect) land-use change and trade-offs between sequestering carbon and replacing

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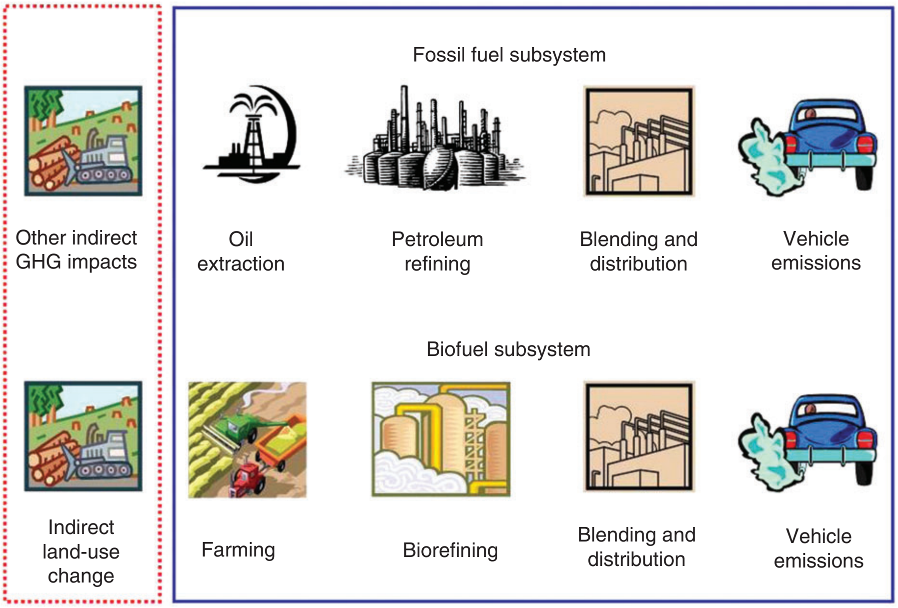

FIGURE 5 | System boundaries as defined for terrestrial resource analysis.

fossil fuels.[51,79] This review did not find TRA studies that specifically analyzed the biofuel systems analyzed in FCA studies; making such quantitative comparisons is a worthy future research task. Nevertheless, it seems likely that the results would differ greatly for biofuels currently in production because TRA does not automatically negate biogenic CO_2 emissions from fuel processing and end-use.

GHG Inventory Accounting

The 'Kyoto accounting' method used for official GHG inventories is geographic in scope.[5] It addresses all GHG sources and sinks in a region (state, nation, geographic region, or entire world), generally without regard to the linkages among products produced or consumed in different regions. The resulting inventories are reported on an annual basis, and so can be considered as having a temporal boundary of 1 year. Although specific inventories are regional, the Kyoto approach presumes a globally complete accounting system overall.

An exception to the lack of linkage between production and consumption is the accounting convention of treating bioenergy as carbon neutral by excluding the biogenic CO_2 emitted in energy sectors. The official guidance is that 'CO_2 emissions from biomass combustion are not included in national totals, but are recorded as an information item for cross-checking purposes as well as avoiding double counting' (IPCC 2:1.19).[5] With CO_2 uptake in feedstock being taken for granted, carbon stock releases due to land-use change are handled in Land Use, Land-Use Change, and Forestry (LULUCF) sectors.[82] Thus, for biofuels, Kyoto accounting embeds a lifecycle perspective in that biofuel use generates a full CO_2 reduction credit in the transportation sector, regardless of the extent to which the assumed carbon neutrality is undone by CO_2 releases in other sectors. The resulting accounting error undermines the environmental integrity of mitigation policies defined on this basis.[42]

Treating all sources and sinks *in situ*, i.e., tallying them in the sectors where positive or negative CO_2 flows actually occur, would address the problems that arise when treating biofuels as carbon neutral in FCA and Kyoto accounting. An approach termed annual basis carbon (ABC) accounting is similar to existing inventory methods except that it includes rather than excludes biogenic emissions in energy sector totals.[83] Fuel emissions are evaluated by chemical carbon content; lifecycle accounting is unnecessary because supply chain emissions and any associated CO_2 uptake are counted in the sectors where they actually occur.

As can be seen in Table 1, end-use emissions vary little among liquid fuels that readily substitute for one another. Therefore, under ABC accounting, no significant CO_2 reduction is attributed to biofuels downstream in end-use sectors. Any such reduction is based on additional net CO_2 uptake upstream in locations of feedstock production. For policy purposes, the implied carbon credit could be calculated using rules similar to those for carbon offsets.[42] For analysis purposes, an independent check

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TABLE 1 | CO_2 Emission Factors and Related Properties of Liquid Fuels

Fuel	Density kg/liter	Carbon fraction	HHV MJ/kg	HHV MJ/liter	LHV MJ/liter	Emissions gCO_2/MJ
Bunker fuel	0.9912	86.8%	42.21	41.84	39.12	80.6
Biodiesel	0.8879	77.6%	40.17	35.66	33.32	75.8
Diesel	0.8469	87.1%	45.77	38.60	36.09	74.9
Crude oil (average)	0.8467	85.3%	45.54	38.56	36.14	73.3
Jet fuel	0.8020	86.2%	46.20	37.05	34.65	73.2
Gasoline	0.7447	86.3%	46.54	34.66	32.36	72.8
Ethanol	0.7893	52.2%	29.85	23.56	21.27	71.0
Butanol	0.8097	64.9%	37.33	30.23	27.83	69.2
Methanol	0.7941	37.5%	22.88	18.17	15.96	68.4

HHV = higher heating value; LHV = lower heating value; emission factor given on a LHV basis.
Source: Fuel properties table from GREET, 2011 edition.[50]

on the extent of net CO_2 uptake could be obtained through techniques for evaluating regional scale net primary productivity (NPP). Such methods have been used to estimate bioenergy production constraints based on NPP[84] and might be extended to generate estimates of additional CO_2 uptake over multiyear periods.

Using ABC accounting for GHG inventories would indicate that CO_2 emissions from liquid fuels must be offset in a generalized sense of the word, a conclusion that also follows from carbon balance analysis.[6] Leakage (including ILUC) remains an issue for any significant effort to offset CO_2 emissions through biomass growth. Nevertheless, leakage might be constrained enough to have confidence in levels of mitigation estimated while monitoring and managing CO_2 uptake based on techniques of terrestrial carbon management.[85,86]

Integrated Assessment Modeling (IAM)

IAM methods jointly model climatic effects and the global economy, accounting for all GHG sources and sinks, their relationships, and the effects of technology and behavioral changes for mitigating or adapting to climate change.[87,52,88] Full or partial equilibrium economic models might be used, but the method always attempts to analyze the world as a whole, including simplified climate models calibrated to detailed geophysical models.[89] IAM has been used to investigate many aspects of global change and to generate scenarios based on economic assumptions and mitigation options, including prospective analyses of large-scale bioenergy systems.[21]

IAM is fully consequential by construction. It treats systems at higher levels of aggregation than does FCA, addressing sources and sinks at regional and sectoral levels, and has not been used to specify fuel regulations. Its complete economic and carbon cycle representations make IAM useful for analyzing policy impacts and informing discussions of mitigation options. However, because its scope is so broad and its results depend on the particular models used, the underlying data and many assumptions, including assumptions about future technologies and behavior, IAM also cannot be verified and must be interpreted carefully. But because it is clearly understood to be a scenario tool, IAM is not prone to misapplication as has been seen with FCA.

A number of IAM studies identify a role for biofuels, including advanced options such as bioenergy with carbon capture and sequestration (BECCS), in long-term climate stabilization, but with clear caveats.[90,91] Attention to the IAM scenario specifications reveals that the extent of bioenergy (including biofuel) use depends greatly on assumptions about technology and land use.[21,92] Results vary according to how the modeling is done, but most studies find significant benefits only for advanced pathways (e.g., widespread availability of cellulosic conversion technologies), increased agricultural yields, minimal countervailing effects (such as N_2O emissions), and sound terrestrial resource management including land-use governance and carbon stock protection.[46,91] IAM also indicates that biofuels offer benefits mainly during the latter half of the 21st century when technology, land use, and other sustainability considerations are taken into account.

Thus, IAM suggests greater caution about biofuels deployment than does FCA. Like TRA, IAM does not suffer from the accounting errors that occur in FCA, Kyoto accounting, or other methods that embed an assumption of biomass carbon neutrality. By

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construction, IAM accounts for leakage effects such as ILUC because all land use-related emissions are modeled. In contrast, and although some researchers sounded warnings,[93,94] policy applications of FCA tautologically assume that biofuels are inherently carbon neutral, resulting in a misplaced burden of proof for CO_2 reduction.[83] Even when enhanced by consequential analyses to handle ILUC, FCA methods rationalize near-term promotion of biofuels by generating CI values that credit decades of assumed future CO_2 uptake against large current-period releases.

A rework of FCA methodology to correct the treatment of baseline land use might yield results more consistent with those of IAM, but would still incur large uncertainties and involve *ad hoc* modeling. Although it may offer some insights, FCA is inferior to scenario analysis using IAM, which has a sound scientific and economic foundation. A leading analyst who pioneered FCA has now reached a similar conclusion.[95]

FCA's prominence in energy policy was secured primarily because of its perceived value for comparing biofuels with each other and with petroleum fuels. This role is seen in the fuel provisions of the RED, the RFS, and California's LCFS, all of which were largely rationalized as ways to expand the use of renewable fuels (*de facto* biofuels). For even longer, FCA has been used to guide research, development, and demonstration programs designed to create renewable replacements for petroleum fuels. Although the method is also used to assess nonliquid alternatives such as electricity, hydrogen, and natural gas, those applications are generally less problematic, at least if biomass is not a feedstock. However, neither is FCA considered very important in those arenas, where it is clear that climate mitigation should target GHG sources upstream rather than, say, trying to control power sector emissions by regulating the lifecycle of electricity delivered downstream through distribution transformers.

CLARIFYING THE PROBLEM STATEMENT

Because liquid fuel-related carbon stocks and flows must be handled dynamically, FCA's static product focus invites an ill-posed question. Asking to compute a CI ('carbon footprint') involves treating an abstract notion—a fuel's lifecycle—as if it were a well-defined fuel property. It is an example of the logical pitfall that Alfred North Whitehead has termed the fallacy of misplaced concreteness.[96] The fuel comparison question as posed through FCA is not a question that can be unambiguously answered; that is to say, it is scientifically irreducible.

On the other hand, well-grounded methods such as IAM may not seem to offer guidance of the type that policymakers seek (the prescription that all carbon should be priced notwithstanding). A way forward can be found by casting the problem in a different light. Figure 6 shows a logical decomposition of mitigation options for motor vehicle CO_2 emissions; similar logic applies to other uses of liquid fuels such as aircraft. This type of analysis is related to the common 'three-legged stool' factorization of transportation GHG emissions into travel demand, vehicle efficiency, and fuel CI.[97] The binary logic of Figure 6 further unbundles the ways to address fuel CO_2 impact, providing a concrete breakdown of options that otherwise are obscured by the LCA abstraction of fuel CI.

The first two branches in Figure 6 represent the level of fuel consumption and the CO_2 emissions impact of the fuel consumed. Fuel consumption depends on travel demand and the fuel use rate of vehicles serving that demand. Those branches interact and are influenced by urban and regional form, infrastructure and mode choice, vehicle fleet mix and fuel efficiency, and behavioral responses. These energy-related factors can also be influenced by the type of fuel; for example, electric cars may be more efficient than conventional vehicles but can have different usage patterns. Such issues are topics unto themselves and not the focus here.

As commonly applied, FCA reduces the CO_2 impact of the fuel to a CI number, creating the problems described above. But as illustrated in the lower branches of Figure 6, whether or not a fuel contains carbon is a clear physical distinction. Noncarbon fuels include energy carriers such as electricity or hydrogen. The fact that electricity and hydrogen do not carry carbon to vehicles is a key reason why extensive efforts are underway to develop technologies for their use in transportation, which is also a topic unto itself.

For a carbon-based fuel, one can either capture the carbon on board the vehicle to avoid releasing CO_2 or counterbalance the vehicle emissions by removing CO_2 from the atmosphere elsewhere. Because on-board carbon capture is not plausible,[98] the only option is removing CO_2 from the atmosphere in locations outside the transportation sector. Biofuels fall under this category because carbon is fixed during feedstock growth. For a climate benefit, however, the amount of CO_2 absorbed from the air must be greater than whatever is already being absorbed through existing activities.[6] This is what is meant by 'net uptake' in that branch of Figure 6. Although it might increase CO_2 removal, sourcing fuel from

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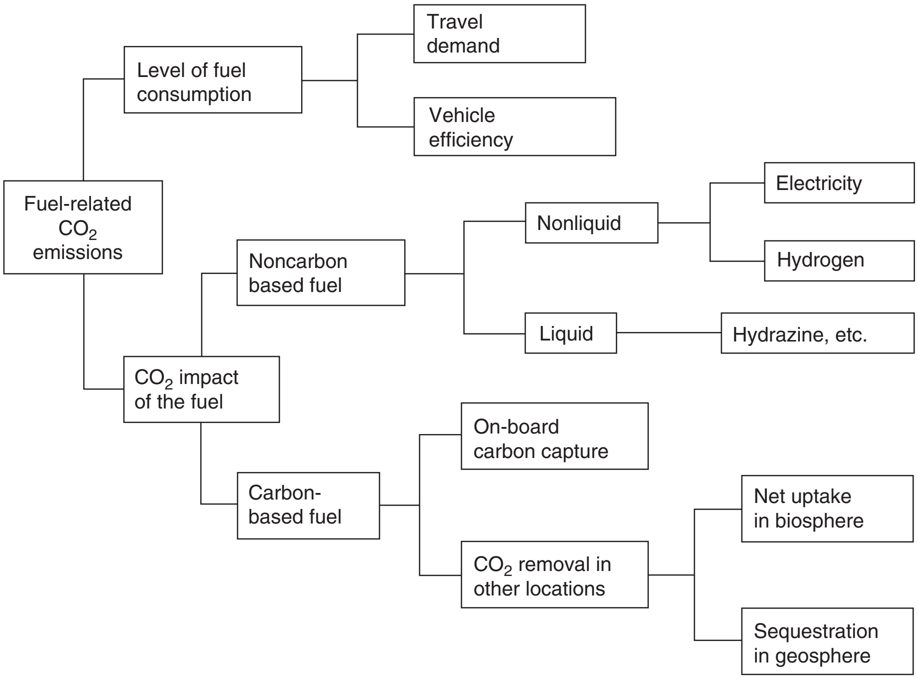

FIGURE 6 | Logic tree for options to address CO_2 emissions from liquid fuel use.

biomass is neither a necessary nor sufficient condition for counterbalancing the CO_2 emitted during fuel end-use. Also, because NPP is the basis for CO_2 removal through biomass production and conversion of land to agriculture often reduces NPP unless significant inputs are provided,[99] an expansion of biofuels production must be balanced against the preservation of primary productivity in the biosphere.

More broadly, the need to counterbalance the CO_2 emitted from the use of liquid fuels points to the need for carbon dioxide removal (CDR) strategies. IAM highlights the importance of CDR for transportation because the sector's mitigation options are projected to be more costly than those for stationary sectors.[100] CDR includes any mechanisms for achieving a net drawdown of carbon from the atmosphere, or 'negative CO_2 emissions' as it is sometimes called.[101] Such options include recarbonization of the biosphere[85] and BECCS[102] as well as chemical direct air capture (DAC) of CO_2,[103] enhanced CO_2-based mineralization, ocean fertilization, and other forms of geoengineering. CDR studies are attuned to the importance of carbon *stock* management (engaging the atmosphere with the biosphere and geosphere) rather than treating mitigation as an emissions *flow* problem as generally done by energy analysts. In general, it is crucial to evaluate net CO_2 removal effects when engaging systems, such as bioenergy, that involve both positive (source) and negative (sink) flows.

RESEARCH AND POLICY IMPLICATIONS

This review critically examined methods for evaluating the net CO_2 impacts of liquid fuels but did not compare their results numerically, which would be a useful next step. Given recent regulatory and market trends, data are becoming available for biofuel and fossil fuel systems at commercial scales, enabling comparisons based on real-world operations rather than assumptions about hypothetical fuel systems. Several types of analysis come to mind:

- Deconstructing FCA models, such as those developed for the RFS and LCFS, to examine spatially and temporally explicit intermediate results. Such analysis would project CO_2 stocks and flows sector by sector in ways that can be compared to inventory-based estimates.
- Developing independent empirical checks on fuel-related carbon accounting and modeling results. Approaches such as those used to assess regional and global changes in net primary productivity and net ecosystem exchange may be useful in this regard.
- Applying TRA methods to actual commercial-scale ethanol and biodiesel production. Although it would not address ILUC, rebound or other induced effects, such analyses

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could bound net CO_2 reduction benefits over time.

- Using IAM with databases sufficiently disaggregate to calculate the impacts of bioenergy systems as actually deployed at commercial scales and under real-world conditions (vs modeling hypothetical systems with stylized land use and technology assumptions).
- Examining the applicability of system dynamics methods. Because fuel systems that engage the biosphere involve stock-and-flow interactions, methods designed for dynamic analysis may be able to represent carbon cycle and economic effects effectively.
- Systematically evaluating current and proposed GHG accounting methods according to intended application (e.g., compiling emissions inventories, analyzing policy options, guiding R&D, specifying regulations, or developing voluntary programs).

Given this range of topics, it could be useful to develop a forum for engaging the different research communities involved and planning the cross-disciplinary coordination needed.

Policy Implications

Energy market trends imply that the need to replace petroleum is less urgent than the need to mitigate climatic risk.[1] Seen in that light, the preceding discussion points to public policy (including R&D) priorities different than those that have animated most work on transportation energy issues to date. Beyond measures to reduce liquid fuel demand, the carbon mitigation problem for liquid fuels reduces to a problem of increasing the rate of net CO_2 removal from the atmosphere.

This perspective suggests greater research emphasis on CDR mechanisms rather than on biofuel synthesis *per se*. Although biofuels can have a role, the first task is securing the additional fixed carbon on which the climate benefit of any biofuel depends. The challenge is to develop ways of removing CO_2 from the atmosphere at faster rates and larger scales than is accomplished by established agricultural and forestry activities; the task involves not only raising productivity (higher yields) but also managing land with carbon in mind. At present, photosynthesis remains the CO_2 removal mechanism to beat, and so both R&D and policy should focus on ways to make the best use of whatever additional fixed carbon becomes available.

Terrestrial biomass-based CDR appears limited to levels well below those needed to offset CO_2 fully from liquid fuels.[104] Nevertheless, meaningful mitigation can be found through reforestation and other ways to recarbonize the biosphere, and so it will be useful to tie bio-based CDR into policies targeting transportation. Affected industries (e.g., petroleum, automotive, freight, and air transport) may find such strategies more cost-effective than many options already under consideration. Alternative vehicle and fuel technologies, for example, have high costs and uncertain benefits.[98] Although bio-based CDR might be criticized as 'just doing offsets' as opposed to being a 'technology solution', the foregoing discussion implies that offsetting is in fact *the* key mitigation mechanism for CO_2 from liquid fuels. Because carbon-based liquids are responsible for anthropogenic emissions of ~3 PgC/year and projected to remain a dominant source of energy for several decades, identifying and pursuing effective strategies for balancing those emissions merits a high priority.

CONCLUSION

Several critical threads emerge when reviewing methods for assessing the CO_2 impact of liquid fuels. One such method is FCA, a form of LCA that has greatly influenced climate-related research priorities and public policies for transportation fuels. Tracking the recent progression of studies, including those undertaken for fuel regulation, and comparing FCA to other methods of analysis, reveals flaws fatal enough to raise serious concerns about the role of FCA in shaping fuel-related CO_2 mitigation strategies.

Given the importance of biofuels as replacements for petroleum fuels, one major concern is that the dynamic stock-and-flow interaction of the biosphere with the atmosphere cannot be reduced to an emissions flow analysis as employed by FCA. This defect is highlighted when comparing FCA to TRA, which is designed to treat the dynamics of biomass resources. Comparing FCA to TRA also reveals a related flaw, namely, the exclusion of existing land use from the reference system baseline. Even though the FCA community has been induced recently (and sometimes reluctantly) to address ILUC, the ensuing *ad hoc* modeling serves only to compound the uncertainties while failing to fix the underlying error of an incorrect baseline.

A second concern is FCA's static framework, which characterizes systems as being in equilibrium when averaged over a predefined lifecycle. The premise is that one such equilibrium system (say,

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a biofuel pathway) can be compared to another (a fossil fuel pathway). However, changing fuel supply systems is a dynamic process, negating the equilibrium premise of FCA. This problem remains even when attributional FCA is overlaid with consequential modeling; because outputs are averaged over an assumed lifecycle, the results still reflect systems analyzed as if they were in equilibrium. In this regard, FCA pales in contrast to IAM, which coherently represents the coupling of economic and climatic systems involved in the dynamics of an energy transition.

Another problem pertains to how FCA fosters discussions that confuse the abstract notion of a lifecycle for a physical reality, a mistake termed the fallacy of misplaced concreteness. In generating what is termed a fuel CI metric, FCA uses the word 'fuel' to represent not just a physical fuel, but rather the multisector industrial systems that supply the fuel. This semantic slippage might not be serious if the systems in question were simple and sufficiently constrained. In reality, liquid fuel supply chains are complex, dynamic, can span the globe spatially and, in the case of biofuels, have impacts that extend into the future temporally.

In treating biofuels as tautologically carbon neutral within their lifecycle, FCA obscures the concrete reality that end-use CO_2 emissions from biofuels differ but little from those of the fossil fuels they replace. Although the carbon in a biofuel was recently removed from the atmosphere, that does not guarantee that it represents a net removal, the extent of which can only be ascertained by quantifying the relevant CO_2 sources and sinks. A similar mistake occurs in the Kyoto convention of treating biomass as carbon neutral. The use of this assumption in two accounting methods that have greatly shaped public policy creates a serious cognitive challenge. Like any fallacy worthy of the label, the grip on reasoning can be quite firm, and so it may take some time before the error is taken to heart by the community of researchers, environmentalists, businesses, and policymakers who are wedded to the lifecycle concept of CI and the methods that propagate it.

Setting the lifecycle paradigm aside clarifies the CO_2 mitigation task for transportation fuels. The liquid carbon challenge is, in fact, a CO_2 removal problem. It requires increasing net CO_2 uptake, in the biosphere or elsewhere, in ways that counterbalance the end-use CO_2 emissions from fuel consumption. An implication is that research should be ramped up on options for increasing the rate at which CO_2 is removed from the atmosphere and on programs to manage and utilize carbon fixed in the biosphere, which offers the best CO_2 removal mechanism now at hand. Such strategies can complement measures that control the demand for liquid fuels by reducing travel activity, improving vehicle efficiency, and shifting to noncarbon fuels. The need for new paradigms to address CO_2 emissions from liquid fuels is an issue that clearly warrants further analysis and discussion. Nevertheless, methods well-grounded in the realities of the carbon cycle may identify mitigation options that are more effective, more accessible, and less costly than those rationalized by FCA and pursued for many years with little meaningful success.

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