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Assessing Resource Intensity and Renewability of Cellulosic Ethanol Technologies Using Eco-LCA

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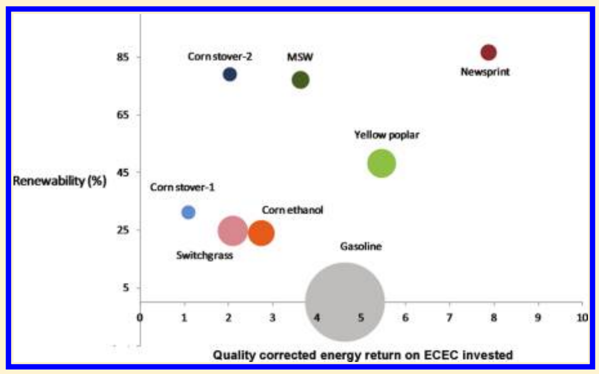
Supporting Information

ABSTRACT: Recognizing the contributions of ecosystem services and the lack of their comprehensive accounting in life cycle assessment (LCA), an in-depth analysis of their contribution in the life cycle of cellulosic ethanol derived from five different feedstocks was conducted, with gasoline and corn ethanol as reference fuels. The relative use intensity of natural resources encompassing land and ecosystem goods and services by cellulosic ethanol was estimated using the Eco-LCA framework. Despite being resource intensive compared to gasoline, cellulosic ethanol offers the possibility of a reduction in crude oil consumption by as much as 96%. Soil erosion and land area requirements can be sources of concern for cellulosic ethanol derived directly from managed agriculture. The analysis of two broad types of thermodynamic metrics, namely: various types of physical return on investment and a renewability index, which indicate competitiveness and sustainability of cellulosic ethanol, respectively, show that only ethanol from waste resources combines a favorable thermodynamic return on investment with a higher renewability index. However, the production potential of ethanol from waste resources is limited. This finding conveys a possible dilemma of biofuels: combining high renewability, high thermodynamic return on investment, and large production capacity may remain elusive. A plot of renewability versus energy return on investment is suggested as one of the options for providing guidance on future biofuel selection.



1. INTRODUCTION

In recent years, there has been a surge of interest in cellulosic ethanol derived from lignocellulosic feedstocks. In addition to the potential effects of climate change, rising crude oil prices, and national security concerns tied to the dependency on foreign oil, there are other compelling reasons for this interest such as the food–fuel conflict. At present, starch-based ethanol, mainly corn is the primary alternative to gasoline in the U.S. Corn ethanol alone cannot meet the growing demand for liquid transportation fuel due to limited available land and competition for food. Recent studies indicate that first generation biofuels such as corn ethanol and soy biodiesel may induce large greenhouse gas (GHG) emissions via indirect land use changes[1,2] which may negate the GHG mitigation benefit of biofuels. There are other environmental concerns emanating from intensive agriculture practices for growing corn such as eutrophication, acidification, water consumption, and downwind impacts of Bt corn pollen.[3]

In contrast, cellulosic ethanol has less water use, lower greenhouse gas emissions,[4] and lower fossil fuel consumption[5−7] due to low fertilizer, pesticide and fossil fuel inputs in the life cycle. For example, the "well-to-wheel" (WTW) GHG emissions can range from −27.5 g CO_2 eq./MJ to 25.5 g CO_2 eq./MJ depending on feedstocks and methods of production.[4] Moreover, indirect land use GHG emissions would be a nonissue for cellulosic ethanol derived from waste materials such as MSW and agriculture residues. A wide range of feedstocks such as forest thinnings, municipal solid waste (MSW), agriculture residues, grasses, and dedicated energy crops can be used for cellulosic ethanol mitigating the risks of feedstock supply. It is estimated that the U.S. has sufficient land to produce about 1 billion tons/year of biomass[8] which when devoted to ethanol production can meet as much as 30% of gasoline demand.[9]

To develop fuels that have a smaller life cycle environmental impact, it is equally important that researchers and policymakers focus on resource consumption, in addition to climate change and other environmental impacts of fuel production and use. Inclusion of sustainability criteria in low carbon fuel-related

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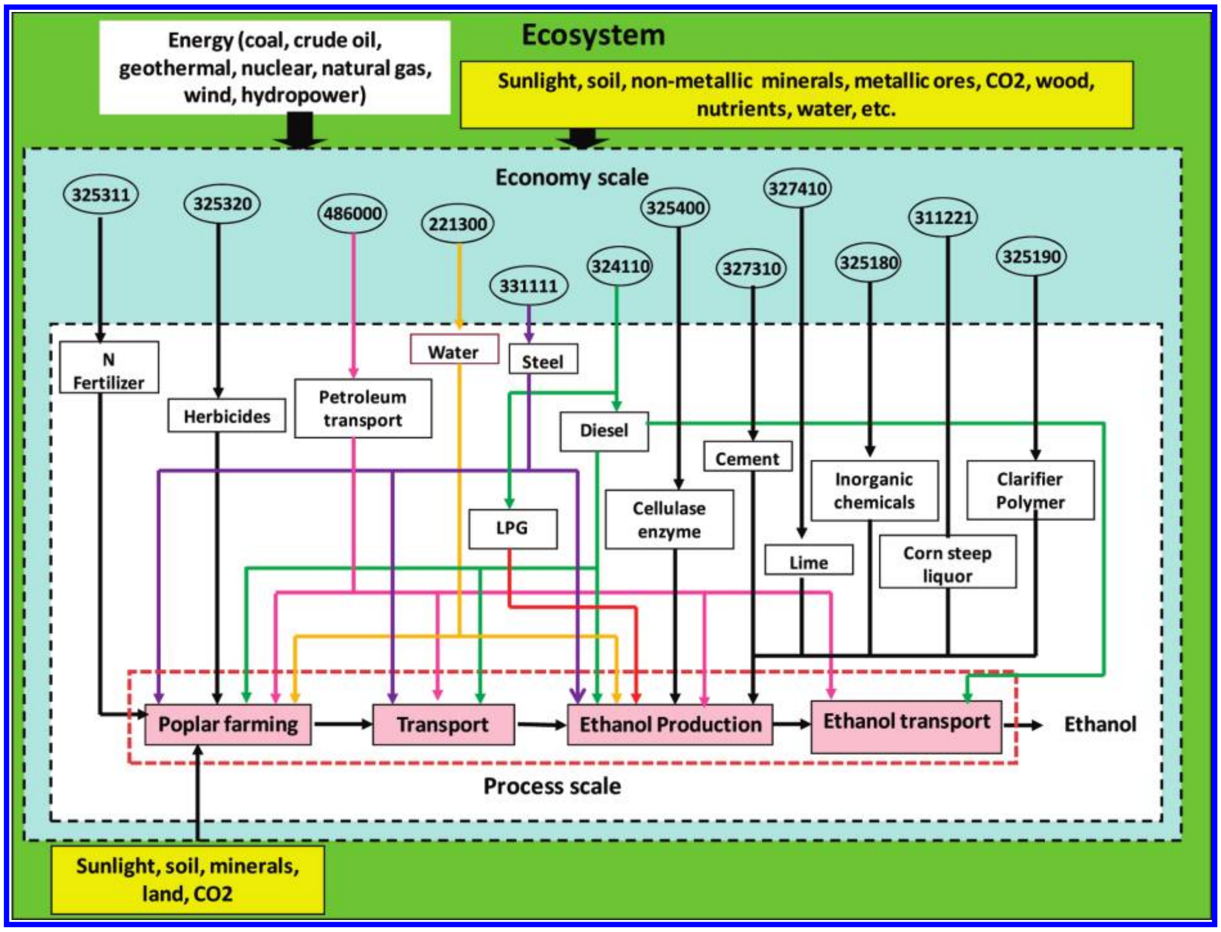

Figure 1. A framework for hybrid Eco-LCA as applied to cellulosic ethanol production from yellow poplar. The numbers in ovals represent NAICS codes for industry.

standards such as the European Renewable Energy Directive and land use restrictions in the U.S. Renewable Fuel Standard (RFS2) reflect an attempt to improve the sustainability of biofuel production by reducing impacts on water, air, biodiversity, and land. A recent emergence of a gamut of voluntary sustainability standards developed by initiatives such as the Roundtable on Sustainable Biofuels (RSB) and Better Sugar Cane Initiative emphasize the resource use aspect of biofuels. The current controversy on indirect land use change (ILUC) and biodiversity has questioned the sustainability of first generation biofuels.[1,2] Therefore, it would be foresighted to reduce the likelihood of unpleasant surprises from large-scale production of new generations of biofuels by including natural resource use in life cycle analysis. Ideally, these resources should include all land and ecosystem goods and services, since without their contribution, human activities cannot be sustained.[10]

Emissions, particularly GHG, and associated environmental impacts of cellulosic ethanol have been studied in detail. Understanding of resource consumption aspects of cellulosic ethanol is quite limited but is important from a sustainability perspective. Existing life cycle inventories focus on non-renewables, including energy resources and emissions but do not account for most renewable ecosystem goods and services such as sunlight, soil, detrital matter, pollination service, fishery, nitrogen and phosphorus mineralization, and nitrogen deposition. Methods like Abiotic Depletion Potential[11] and Surplus Energy[12] focus on aggregating nonrenewable resources, whereas Ecological Footprint Analysis[13] deals with renewable resources. Exergy based LCA has been studied for various fuels,[14,15] but those studies ignore consumption of resources whose exergy cannot be directly quantified such as land and pollination services, and the role that nature plays in making resources available. Relying mainly on aggregate metrics has the disadvantage of losing details. Therefore, a hierarchy of metrics is preferable.[10]

Ecologically Based LCA (Eco-LCA) is a recent life cycle oriented approach that aims to account for the role of natural resources such as land and ecosystem goods and services in LCA.[10] This approach considers a wide array of goods and services derived from nature and a hierarchical, thermodynamic aggregation scheme to permit meaningful interpretation. In this study, Eco-LCA is used to compare various attributes of cellulosic biofuels obtained from five different feedstocks for informing the policy debate: switchgrass, yellow poplar, corn stover, newsprint, and municipal solid waste (MSW). Large-scale cellulosic ethanol production may entail a substantial shift in consumption of resources such as land, water, soil, and minerals. Hence, natural resource consumption needs to be quantified in a scientifically consistent and rigorous manner. Results from Eco-LCA provide insight into the vulnerability of selected fuels to the depletion of specific resources. Aggregate quantities such as a quality corrected thermodynamic return on investment and renewability index help in gauging the sustainability and resource competitiveness of the selected cellulosic fuels with respect to gasoline. These aggregate metrics

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Table 1. Feedstock and Ethanol Yields for Various Lignocellulosic Feedstocks

feedstock yield (dry MT/ha)		corn stover[26]	yellow poplar[27]	switchgrass[8]	MSW	newsprint
	high	8.5	13.9	22.7		
	default	7.7[a]	13.1	16.0		
	low	6.8	10.0	9.4		
ethanol yield (liters/dry MT)[b]		corn stover	yellow poplar	switchgrass	MSW	newsprint
	high	353	365	340	267	421
	default	340	352	327	257	405
	low	255	264	245	192	303

[a]Calculated based on the corn grain yields/ha.[26] It was assumed that corn stover and corn seeds are produced in 1:1 ratio, and the moisture content of corn grains is 15%. To minimize soil erosion and maintain soil fertility, only 62% of corn stover produced is harvested.[28] [b]Ethanol yields of switchgrass, poplar, newsprint and MSW are calculated in reference to the yield of corn stover considering cellulose and hemicellulose contents. The default corn stover yield was taken from Sheehan et al.[25] It was assumed that 90% of cellulose and hemicelluose are converted to sugars and conversion rates for glucose sugar and pentose sugars are 95% and 85%, respectively.

are based on the concept of Ecological Cumulative Exergy Consumption (ECEC)[16] which is closely related to emergy.[17] ECEC adjusts for qualities of resources being aggregated, enhancing the value of energy return on investment metrics, and provides a balanced indication of renewability.[17] The issue of quality adjustment of resources has received attention especially in the context of energy analysis in LCA of biofuels, with more details elsewhere.[18−20]

This study focuses on three main aspects of sustainability related to biofuels: (1) resource intensity, (2) energy competitiveness, and (3) renewability. Ideally, future fuels should be energetically competitive, renewable, and available in large enough quantities to make a substantial dent in satisfying our transportation fuel requirements. This study demonstrates the dilemma facing biofuels since high return on energy invested, high renewability, and large quantities seem difficult to achieve simultaneously.

2. MATERIALS AND METHODS

2.1. Models. Cumulative resource consumption from "well-to-tank" for various fuels was estimated by taking into account economic and natural resource inputs along the supply chains in Hybrid Eco-LCA models. In this study, a hybrid model was developed by combining an economic input-output life-cycle inventory from Eco-LCA[10] with a process-based inventory for the production and use phases. Eco-LCA uses an environmentally extended economic input-output model of economic sectors, similar to the approach of Economic Input-Output LCA (EIOLCA)[21] except that it considers a wide range of land and ecosystem goods and services in addition to energy and emissions, and uses thermodynamic aggregation metrics. Producer prices were used to calculate resource consumption by economic inputs in well-to-tank of fuel life cycles by inputting them in the Eco-LCA model built on the well-known Leontief input-output equation. The lists of economic inputs, their prices, and direct inputs of natural resources in farming are provided in the Supporting Information (SI) (Tables S11−S20). An illustrative framework of a hybrid Eco-LCA model for cellulosic ethanol produced from yellow poplar is shown in Figure 1. Details on constructing a hybrid Eco-LCA model are in the Supporting Information of Baral et al.[18]

Eco-LCA[22] is a versatile tool for quantification of cumulative consumption of natural resources and emissions over the life cycle once the producer prices of economic products or services are determined. As Zhang et al.[10] describe in more detail, Eco-LCA considers provisioning services such as fossil fuels, minerals, timber, and fish; and regulating and maintenance services such as pollination, nutrient cycling, and carbon sequestration by associating them as inputs to (demand side approach) or outputs from (supply side approach) the related industry sectors. For example, iron ore is considered as an input from nature to the iron ore mining sector from where it flows into other industry sectors such as iron and steel mills. Coal is also an input to coal mining or its use may be estimated by knowing the electricity used by various industry sectors. Eco-LCA allows one to normalize resource consumption/flows with national resource consumption or flows to identify vulnerabilities and constraints for a given level of production of a product such as biofuel. More importantly, natural resources can be aggregated in terms of mass, energy, and exergy to elicit complementary information about the product being studied.

2.2. Data Sources and Assumptions. Five different lignocellulosic feedstocks were chosen to represent variation in chemical compositions of feedstocks and methods of cellulosic ethanol production through chemical and biochemical routes. For switchgrass, yellow poplar, corn stover and newsprint, cellulosic ethanol was considered to be produced using acid pretreatment followed by a simultaneous saccharification and fermentation (SSF) process,[23] whereas the Gravity Pressure Vessel method[24] was assumed for cellulosic ethanol production from MSW. Since gasoline is the target fuel for substitution by biofuels, the life cycle assessment of gasoline is provided as a reference. First tier (direct) inputs such as materials, chemicals, and energy were quantified assuming optimistic ethanol yields (default values) as shown in Table 1. Energy and chemical requirements for ethanol production from corn stover were based on Aden et al.[23] and were adjusted for optimistic yield of 240 L/dry metric tonne (MT).[25] Chemical requirements (sulfuric acid, lime, and cellulase enzymes) for yellow poplar and switchgrass were derived by modifying corn stover data by taking into account the difference in hemicellulose and cellulose contents. As an approximation, amounts of other chemicals such as wastewater treatment and process water were assumed to be the same as that of corn stover per liter of ethanol produced. The process input data for newsprint were projected from Kemppainen and Shonnard[29] considering an optimistic yield of 405 L and assuming that an increase in yield results in a linear decrease in energy and chemical consumption. Similarly, for MSW, process input data provided by Kalogo et al.[24] was projected to reflect an increase in yield. They assumed a yield of 84 L/wet MT MSW, whereas a yield of 229 L/wet MT (257 L/dry MT) was considered in this study as a best case scenario for cellulose and hemicelluloses conversions, which is in line with similar scenarios for other feedstocks.[25,28] Energy expended to deliver chemicals to ethanol facilities and required to transport fuels from the point of production to distribution centers were taken from GREET.[30] Data about steel and cement used in ethanol production facilities were adopted from Felix.[31] Although energy and chemicals required to treat wastewater from ethanol plants were modeled, other air and water pollution control measures such as runoff treatment were not modeled. For example, buffer strips and rural land conservation policies are used frequently and successfully to mitigate runoff impacts.

2.3. Data Organization. *2.3.1. Disaggregated Data.* Life cycle use of individual natural resources for cellulosic ethanol obtained from a hybrid Eco-LCA were compared to that of gasoline to assess each fuel's relative resource intensity. Resource consumption can be normalized with respect to U.S. flows/consumption to assess resource constraints and vulnerabilities for large-scale cellulosic ethanol production. Disaggregate data help present details about resources including those that are difficult to aggregate (e.g., land and pollination services) by the selected physical methods.

2.3.2. Aggregated Data. Consumption data of individual resources were further aggregated in this study using energy analysis as shown below.

$$E = \sum x_i \tag{1}$$

Here, E refers to cumulative energy consumption and x_i represents the calorific value (joules) of the ith resource. This aggregation scheme implicitly assumes substitutability of energy inputs, which needs to be done carefully with cognizance of differences in resource quality. Similarly, industrial cumulative exergy consumption (ICEC) can be calculated using eq 1 when x_i represents the useful energy (exergy) of the ith resource. Unlike energy analysis, ICEC analysis allows us to aggregate material and energy inputs on a common basis of available useful energy. To overcome the shortcomings of energy and exergy analyses, Ecological Cumulative Exergy Consumption (ECEC) has been proposed as a concept that can account for quality differences of resources consumed.[16] ECEC or emergy[17] analysis attempts to correct qualities of resources by multiplying physical characteristics such as energy, exergy, or mass with a quality correction factor known as transformity (τ_i). ECEC is measured in solar equivalent joule (sej).

$$\text{ECEC} = \sum x_i \times \tau_i \tag{2}$$

Here, x_i represents the physical value such as energy, exergy or mass of the ith resource (fuel or nonfuel) and τ_i is the transformity for the corresponding resource. Transformity (τ_i) is defined as the total work required (sej) to produce a unit (mass, energy, exergy) of a product. One shortcoming of ECEC analysis is uncertainty in transformity values due to consideration of system level energy and material flows. Each of these approaches, limitations notwithstanding, can provide unique insight into life cycle analysis and are useful in informing science and policy.

2.4. Aggregated Metrics. *2.4.1. Return on Investment.* Two types of return on investment metrics were calculated: (1) energy return on energy investment (r_E), and (2) quality corrected exergy return on emergy (ECEC) investment (r_{Em}). Energy return on energy investment has been the most used metric to compare energetic competitiveness of alternative fuels and accounts for only fuel value. It is defined as the ratio of output energy (E_o) to process energy (E_p).[32]

$$r_E = \frac{E_o}{E_p} \tag{3}$$

E_o is the sum of the energy content of a product and the energy credit from coproduct electricity from lignin combustion and/or energy saved from land filling. Energy credit was calculated using market value allocation and displacement method as discussed later in this section. Process energy (E_p) refers to the energy consumed in processing the feedstock to fuel, and does not become a part of the product energy. For example, electricity and steam produced from lignin and used in an ethanol plant are part of process energy whereas hemicellulose and cellulose converted to ethanol are not.

By considering only nonrenewable process energy (E_{pn}), we calculate a variation of r_E as,

$$r_{En} = \frac{E_o}{E_{pn}} \tag{4}$$

For cellulosic ethanol, E_{pn} excludes energy obtained from lignin combustion since it is considered to be renewable. r_{En} indirectly represents the degree of fossil fuel savings through gasoline or diesel displacement. For example, if a fuel can be produced with little fossil input (i.e., large r_{En}), fossil fuel savings would be large. r_E and its variation r_{En} deal only with process energy inputs and reflect how efficiently the desired energy output can be generated per unit of process energy (or nonrenewable process energy for r_{En}). These metrics do not consider the quality of energy and nonfuel inputs.

Similarly, quality adjusted energy return on emergy or ECEC investment (r_{Em}) is defined as the quality adjusted output exergy divided by ECEC of the inputs purchased from the economy for processing the feedstock to fuel. These purchased processing inputs, F, include energy, materials, and services. This indicator shows how much quality adjusted useful energy can be generated per unit of work (sej) done by the economy.

$$r_{Em} = \frac{E_x \times \tau_{max} + Em_{credit}}{F} \tag{5}$$

Quality adjusted output exergy is the sum of the quality adjusted exergy content of a product ($E_x \times \tau_{max}$) and emergy credit from a coproduct (Em_{credit}). τ_{max} is the maximum transformity among fuels studied for the same usefulness or functional unit, which in this study is gasoline. Including τ_{max} makes the results more intuitive by adjusting the quality in terms of solar energy equivalent (sej) of the produced fuel to make it comparable with other fuels.[33]

2.4.2. Renewability Index. Renewability index is an important metric based on the relative contribution of renewable resources in production systems.[18] Renewability index (%) is given as:

$$\text{renewability index}(\%) = \frac{R}{T} \times 100 \tag{6}$$

where R is renewable ECEC and T is total ECEC. Here, natural resources are classified as being either renewable or nonrenewable, but more sophisticated definitions could also be used. Renewability calculated from ECEC provides a balanced measure of renewability by recognizing the quality differences among sunlight, fossil fuels, and other natural resources.[18]

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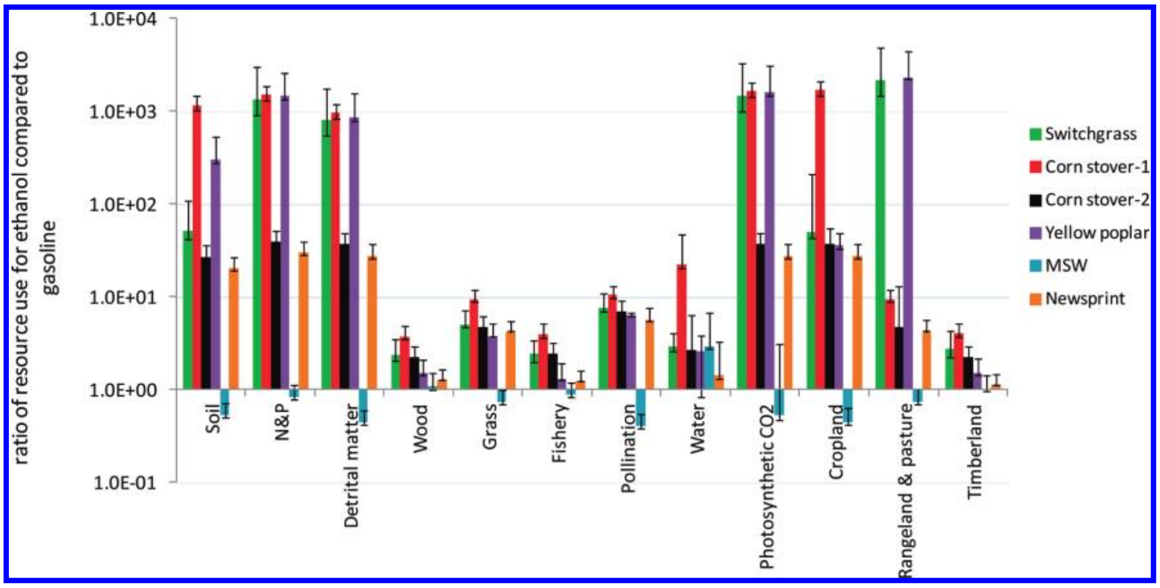

Figure 2. Relative consumption/use of land and various ecosystem goods and services for cellulosic ethanol with respect to gasoline on a per km basis, modeled after a 2006 Chevrolet Impala, with market value-based allocation (log scale). Error bars represent the range obtained from sensitivity analysis. Graphs for other resources are in the SI as Figures S1 and S2.

2.5. Allocation. Two allocation scenarios were considered for corn stover. In scenario corn stover-1, corn stover was considered as a valuable byproduct and agricultural inputs were allocated equally between corn and corn stover on a mass basis, while in scenario corn stover-2, corn stover was considered as waste and all agricultural inputs were allocated to corn. The corn stover-1 case was included as an allocation scheme that assigned a large fraction of the impact to stover, making it a worst case scenario.

Except for MSW, lignin (a byproduct of cellulosic ethanol production) was assumed combusted to generate steam and electricity for in-plant use and selling to the grid. Hence, excess electricity was considered as a coproduct. Allocations were done in two ways: (1) market value-based allocation and (2) displacement method. In the displacement method, it was assumed that excess electricity displaces the electricity produced from the power generation sector. The electricity displaced represents the national 1997 electricity mix, and the electricity price and Eco-LCA were used to determine the resources avoided due to excess electricity. For MSW and newsprint, diverting waste from landfill to an ethanol facility avoids energy (diesel, gasoline, and electricity) use in landfilling and considered for allocation. Unless otherwise indicated, results presented here are based on market value-based allocation. For comparison purposes, results obtained from the displacement method are provided in the SI.

2.6. Sensitivity Analysis. Since cellulosic ethanol technology is still in its infancy, uncertainties are present in the relevant production models. To capture the uncertainties, sensitivity analysis was performed by varying the feedstock yield per ha and ethanol yield per dry MT biomass. A combination of a low feedstock yield/ha and a low ethanol yield/dry MT reported in the literature was chosen to represent a conservative scenario, whereas a combination of a high feedstock yield and a high ethanol yield was chosen to represent the optimistic near term future scenario. High and low yields along with default values are presented in Table 1. The basis for choosing low and high ethanol yields is as follows. According to Spatari et al.[28] high ethanol yield for switchgrass for the year 2010 is 340 L/dry MT which represents +3.8% of the default value. For corn stover, the conservative estimate of yield is 255 L/dry MT,[25] which is −25% of the default value. Therefore, high yields were taken to be +3.8% of the default value and low yields were taken to be −25% of the default value for all fuels. For feedstock yield/ha, the range suggested in the literature was considered to define low and high yields.[8,26,27]

3. RESULTS

3.1. Disaggregated Data. Ratios of individual ecosystem goods and services and land used/consumed by cellulosic ethanol to that of gasoline provide an indication of relative resource intensity of cellulosic ethanol for comparing alternative fuels.

3.1.1. Relative Resource Intensity. Eco-LCA provides information about relative consumption/use of an array of land and ecosystem goods and services. Some of these are shown in Figure 2 for the case when cellulosic ethanol replaces gasoline on a per km basis. Similar graphs for other ecosystem services are in the SI as Figures S1 and S2. In these figures, a value less than 1 suggests that cellulosic ethanol substitution would reduce consumption/use of a particular resource or service with respect to gasoline and a value greater than 1 suggests that cellulosic ethanol substitution would increase its use. The life cycle resource consumption data for gasoline used in deriving the ratios were also obtained using a hybrid Eco-LCA approach described in Section 2.

Crude oil consumption varies from 1.9 MJ/gasoline eq. liter (GEL) for MSW to 8.2 MJ/GEL for corn stover-1. These figures are comparable to crude oil consumption by cellulosic ethanol obtained from herbaceous biomass and farmed trees as reported in GREET.[30] In this study, a gasoline eq. liter is based on the capacity to drive a 2006 Chevrolet Impala for a kilometer. This represents a reduction in crude oil consumption in the life cycle by a factor ranging from 7 to 28 when cellulosic ethanol displaces gasoline (SI Figure S1). This would translate into reductions of about 88−96% of crude oil consumption in the life cycle. It suggests that if a primary goal is to reduce dependence on imported crude oil, cellulosic ethanol can be a

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fuel of choice. On the other hand, cellulosic ethanol consumes more natural gas and electricity (for some feedstocks) relative to gasoline (SI Figure S1). In terms of metallic ores, sand, and crushed stone, cellulosic ethanol is more resource intensive than gasoline (SI Figure S5), but as normalized data in the SI (Figure S4) show, their use is only a small fraction of the national use of these resources. Similarly, cellulosic ethanol consumes more ecosystem goods and services including soil, nutrients, detrital matter, water, and pollination service than gasoline (Figure 2).

Depending on feedstocks, water use by cellulosic ethanol is 1.4–2.9 times more than gasoline, which is 21–30 kg/GEL, and when corn stover is not considered waste (corn stover-1), it is 22 times more than gasoline. Water use refers to both consumptive use such as in irrigation and nonconsumptive use such as in electricity production. For a reference, Wu et al.[34] reported consumptive water use for switchgrass ethanol in the range 2.8–14.8 kg/GEL. Land use is high for feedstocks directly dependent on agriculture. For example, pastureland use requirements are 2,140 and 2,260 times more for switchgrass (2.8×10^{-4} ha/GEL) and yellow poplar (2.9×10^{-4} ha/GEL), respectively as compared to gasoline. Cropland use is 36 (6.6×10^{-6} ha/GEL) and 1670 times more (3.0×10^{-4} ha/GEL) for corn stover-2 and corn stover-1, respectively, compared to gasoline. The larger land area requirement for corn stover-1 is because not all corn stover produced can be used for ethanol production. Some of it (about 38%) is left behind for maintaining soil quality. USDA research[35] has shown that corn stover can be removed only when corn yields exceed 62.8 kg/ha. With lower yields, any stover removal depletes soil organic matter and leads to erosion. Note that land use estimates do not include market-mediated indirect land use change. Cellulosic ethanol is associated with significant soil consumption (erosion), especially corn stover-1 relative to gasoline. For switchgrass, soil erosion is limited due to its extensive root system, which strongly binds soil.

3.2. Aggregated Metrics. The underlying objective of aggregate energy and ECEC metrics is to derive additional and complementary insight with regard to cellulosic biofuels. Two types of aggregate metrics were calculated: (1) return on investment, and (2) renewability index, to assess energy competitiveness and renewability, respectively.

3.2.1. Return on Investment Metrics. When total process energy is considered, energy return on investment for cellulosic ethanol ranges from 0.7 for corn stover-1 to 2.7 for MSW (Figure 3) under different allocation approaches. Except for MSW, energy returns are either similar to or lower than that of corn ethanol. Previous studies showed r_E of less than one for cellulosic ethanol derived from dedicated energy crops.[5,30] For MSW, Kalogo et al.[24] found an r_E value of 1.7 when MSW classification was considered in the analysis. Thus, from an energy competitiveness point of view, cellulosic ethanol from different feedstocks does not seem to be competitive to gasoline with r_E of 3.4 (market value-based allocation) and in most cases to corn ethanol. This suggests that improvements in process designs are necessary to improve the r_E of cellulosic ethanol.

When only nonrenewable process energy is considered, energy return on investment (r_{En}) of cellulosic ethanol is either close or higher than that of gasoline (Figure 3), particularly for yellow poplar and newsprint since a large fraction of process energy requirements for ethanol production are met by lignin, and/or energy inputs in farming are relatively small. Before allocation, renewable energy from lignin accounted for 82% of the total well-to-tank energy consumption for yellow poplar. This means cellulosic ethanol can be a good alternative to corn ethanol in reducing dependence on fossil fuels and hence reducing greenhouse gas emissions. In general, the displacement method for allocation resulted in slightly higher r_E values than the market value-based allocation. The lowest r_{En} was obtained for corn stover-1 because in this scenario agricultural inputs for corn production were equally split.

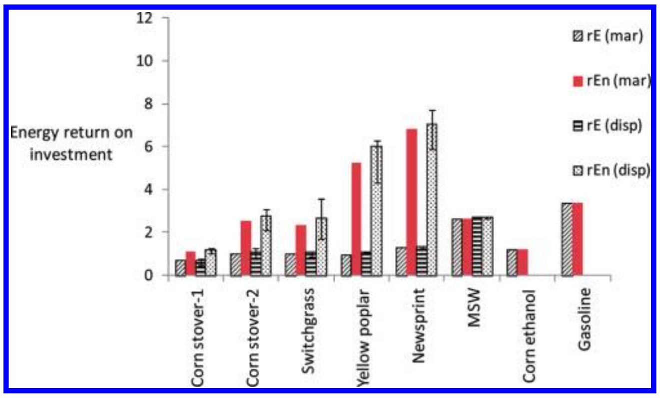

Figure 3. Energy return on investment for cellulosic ethanol, corn ethanol, and gasoline.

For cellulosic ethanol the quality adjusted energy returns on emergy investment (r_{Em}) are comparable or higher than that of corn ethanol, except for scenario corn stover-1 (Figure 4). The higher r_{Em} for yellow poplar and newsprint suggests the economy has to invest less work to produce 1 J of output. As discussed earlier, the major energy input to the process comes from lignin, which in turn comes from sunlight, a direct input from nature. The larger the work provided by ecosystem goods and services, the smaller the effort required from economic goods and services for processing a raw material into a useable product. This relative contribution from nature versus the economy is an indication of product competitiveness and is measured by the quality adjusted energy return.

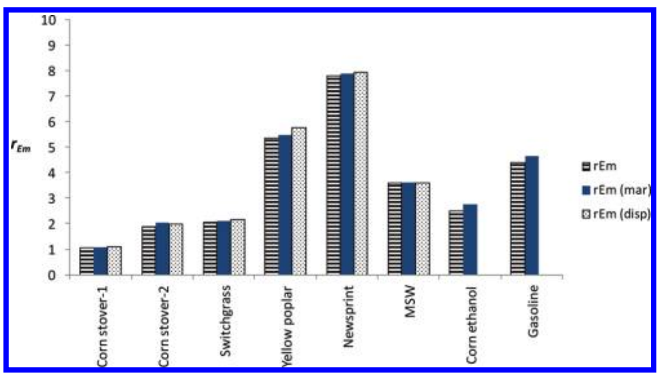

Figure 4. Quality adjusted energy return on ecological cumulative exergy (emergy) investment (r_{Em}) for various fuels.

3.2.2. Renewability, Energy Competiveness, and Production Potential. From the perspective of sustainability and competitiveness, it is desirable that the alternative fuels are renewable and have higher returns on investment. In addition, an ideal alternative fuel should be produced in a large quantity to have a measurable positive impact on energy security and the environment. A fuel that ranks high in these three categories can be considered as an excellent future alternative to fossil fuels. In order to compare various fuels based on these three attributes, Figure 5 shows a bubble chart of the energy return

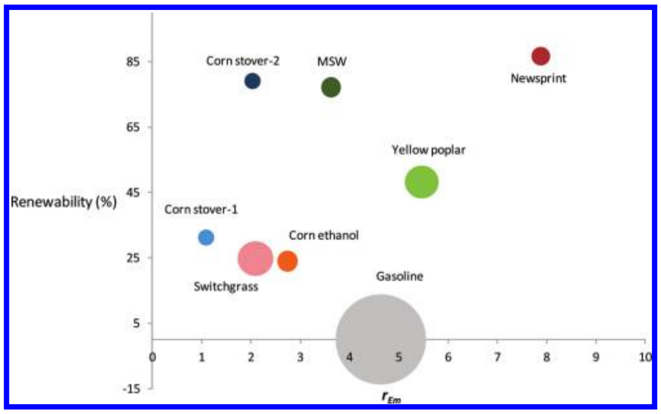

Figure 5. Renewability vs r_{Em} and production potential as metrics for policy guidance. For reference, the size of the bubble for corn ethanol represents 27.9 billion gasoline equivalent liters produced in the U.S. in 2009. One liter of gasoline is equivalent to 1.44 L of ethanol based on its capacity to drive a 2006 Chevrolet Impala for one kilometer.

on emergy investment versus the renewability index (%), with the area of a bubble indicating the production potential. The production potential is theoretical and does not consider competing uses of a given feedstock and land. The bubble size for corn ethanol reflects the gasoline equivalent volume (27.9 billion GEL) produced in 2009 in the U.S. and that for gasoline reflects the volume (522 billion liters) used in 2009, providing reference points for comparison.

Cellulosic ethanol derived from newsprint, MSW and yellow poplar show relatively high renewability and r_{Em}. However, the theoretical biofuel production volumes of these fuels are smaller relative to gasoline consumption in the US due to limited land and feedstock availability. The relatively higher renewability of corn stover-2, MSW, and newsprint is an overestimate since the entire feedstock (waste) was considered to be renewable. This is because it is very difficult to quantify renewable and nonrenewable energy embodied in waste products such as MSW.

3.2.3. Allocation Issue. Allocating energy based on market values of coproducts resulted in lower returns on investment compared to the displacement method with few exceptions. For resource consumption, however, no conclusive trends can be established for the market-based allocation and displacement methods. As the cases of corn stover show, considering a feedstock as waste or a valuable product can have a significant impact on energy and resource intensity. When considered as waste, footprints of cellulosic ethanol improve appreciably. In this study, MSW and newsprint are considered as waste without any embodied energy and resource consumption in analyzing resource intensity and return on investment metrics. If newsprint and MSW were considered as valuable economic products, energy and resources used in making paper or crop cultivation would be allocated to the use of newsprint or organic waste as feedstocks for fuels increasing their resource and energy intensity, but such an analysis is beyond the scope of the study. This is similar to allocating environmental burdens among multiple uses of a product.

4. DISCUSSION

While GHG reduction potential of biofuel, the most common comparison criterion, is important for evaluating biofuels, a number of indicators need to be considered to ensure that biofuels do not shift their environmental impact from GHG emissions to other impacts. This paper offers a comprehensive analysis of cellulosic ethanol in terms of resource use and renewability by considering land use and a number of ecosystem goods and services. The major thrust behind choosing five different feedstocks is to capture the variability in feedstock compositions and production methods to provide a proper perspective about the potential of cellulosic ethanol to be a viable and sustainable fuel alternative.

The disaggregated data analysis reveals that cellulosic ethanol is more resource intensive in terms of consumption of land and a number of ecosystem goods and services than gasoline with few exceptions (for example, crude oil). This is especially so for cellulosic ethanol derived from feedstocks which are directly dependent on agriculture. Cellulosic ethanol from MSW and newsprint consumes less cropland, ores, and minerals, and it causes less soil erosion than cellulosic ethanol derived from other feedstocks. Although this conclusion is intuitive and hence less surprising, quantification of natural resource consumption is one unique aspect of this study. Despite being resource intensive, some of these ecosystem resources are either renewable such as detrital matter, sunlight, and photosynthetic CO_2 or are not consumed in large quantities such as ores and minerals, so they may not pose a constraint for large-scale production. The major constraints for large-scale production can be land use and/or soil erosion for cellulosic ethanol derived from corn stover, switchgrass, and yellow poplar. However, cellulosic ethanol offers the promise of reducing crude oil consumption mainly due to the use of lignin as a source of process energy.

This study highlights that energy return on energy invested (r_E) of cellulosic ethanol is not higher than that of corn ethanol except for MSW and newsprint. An $r_E > 2$ is considered good. What is attractive is the energy return on nonrenewable energy (r_{En}>2), for all cellulosic feedstocks indicating the potential of cellulosic ethanol to generate more energy output per unit of nonrenewable energy input. However, it does not indicate the overall energy competitiveness but rather a potential to reduce fossil fuel consumption. A slightly higher r_E for MSW derived cellulosic ethanol is promising since tipping fees paid for disposal of MSW may lower the cost of production and may well make MSW-based ethanol economically more competitive than ethanol derived from other sources. Tipping fees vary from \$15/MT to \$100/MT in the U.S.[36] which amounts to \$0.25−1.65 per gallon of ethanol produced. Nonetheless, it should be viewed against a possible increase in hazardous waste disposal cost due to accumulation of hazardous waste from MSW processing.

The quality corrected energy return on ECEC invested (r_{Em}) of yellow poplar- and newsprint-derived cellulosic biofuels compare favorably with gasoline indicating they require less work from the economy and may be as competitive as gasoline, and more competitive than corn ethanol. The higher renewability of waste-derived fuels and yellow poplar as compared to corn ethanol emphasizes the value of biofuels that utilize waste products and/or less input of fossil energy.

A graph like Figure 5 can serve as a policy guide for choosing between alternative fuels and improving their environmental performance. If acceptable threshold values were set for renewability and return on investment metrics, the quadrant of more desirable fuels could be drawn. Such methods for decision support may be considered in future work. An alternative analysis could be done with renewability versus r_E or r_{En} graphs (See SI).

When return on investment, renewability and production potential are considered together, a clear pattern of trade-offs emerge. Cellulosic ethanol from waste-based feedstocks, newsprint and MSW, offer decent energy returns considering their r_E in the range 1.3−2.7 (Figure 3) and also excellent returns on emergy with r_{Em} in the range of 3.6−7.2 (Figure 4). These fuels seem sustainable due to a higher degree of renewability (>70%), but they fail to meet a significant portion of the U.S. gasoline demand due to limited production potential. For example, MSW may provide 24.6 billion GEL of cellulosic ethanol compared to the US gasoline demand of 522 billion liters in 2009. Corn stover, despite being a waste product, has lower returns on investment. On the other hand, switchgrass cellulosic ethanol may provide a modest volume of cellulosic ethanol (77.5 billion GEL), but its renewability index is lower, as are the returns on investment- r_E and r_{Em}. Also, its resource intensity is larger among the cellulosic biofuels studied here. For yellow poplar, although renewability (48%) and production potential (71.2 billion GEL) are decent, its r_E of about 1 means it is energetically inefficient, although its r_{Em} is higher.

There is no panacea as none of the cellulosic ethanol fuel systems simultaneously provide a high return on investment, high renewability, and a large production capacity. By looking at the attributes of MSW and newsprint, which require no agricultural inputs but have limited feedstock availability, and yellow poplar which requires less agricultural input, it emerges that the above-mentioned requirements may be satisfied by pooling together feedstocks that can be grown without intensive agriculture on marginal land with good productivity, and waste-based feedstocks and/or improving process efficiency. Policy instruments such as credits for biofuels from wastes and marginal lands, responsible cultivation areas, and sustainability standards may create drivers for achieving this objective. An analysis of impacts of process efficiency improvements and the effects of integrated industrial and ecological designs (such as using wetlands for runoff treatment and biosolids as fertilizers) on resource intensity and sustainability can further enhance the renewability and energy return of biofuels and are areas of future research.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Detailed inventory data, calculations and additional figures are in the Supporting Information. This material is available free of charge via the Internet at http://pubs.acs.org.

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