Life cycle assessment of lignocellulosic ethanol: a review of key factors and methods affecting calculated GHG emissions and energy use

Kelsey Gerbrandt¹, Pei Lin Chu¹, Allison Simmonds¹, Kimberley A Mullins², Heather L MacLean¹,³,⁴, W Michael Griffin⁵ and Bradley A Saville¹,³,⁴

Lignocellulosic ethanol has potential for lower life cycle greenhouse gas emissions compared to gasoline and conventional grain-based ethanol. Ethanol production ‘pathways’ need to meet economic and environmental goals. Numerous life cycle assessments of lignocellulosic ethanol have been published over the last 15 years, but gaps remain in understanding life cycle performance due to insufficient data, and model and methodological issues. We highlight key aspects of these issues, drawing on literature and a case study of corn stover ethanol. Challenges include the complexity of feedstock/ecosystems and market-mediated aspects and the short history of commercial lignocellulosic ethanol facilities, which collectively have led to uncertainty in GHG emissions estimates, and to debates on LCA methods and the role of uncertainty in decision making.

Addresses
¹ Savant Technical Consulting, Toronto, ON, Canada
² University of Minnesota, Department of Bioproducts and Biosystems Engineering, St. Paul, MN, USA
³ University of Toronto, Department of Chemical Engineering and Applied Chemistry, Toronto, ON, Canada
⁴ University of Toronto, Department of Civil Engineering, Toronto, ON, Canada
⁵ Department of Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA, USA

Corresponding author: MacLean, Heather L (heatherl.maclean@utoronto.ca)

Introduction

Lignocellulosic ethanol offers the potential to diversify the transportation fuel pool with a renewable liquid fuel produced from a range of feedstocks, and can lower life cycle greenhouse gas (GHG) emissions intensity relative to conventional ethanol produced from grain and sugarcane. However, to ensure ethanol production ‘pathways’ meet environmental goals, improved understanding of the life cycle environmental impacts of lignocellulosic ethanol is critical before wider scale deployment.

Ethanol produced from corn grain and sugarcane are the dominant alternative light-duty vehicle transportation fuels replacing gasoline. Lignocellulosic ethanol has garnered significant attention over the past 15 years and consequently, numerous life cycle assessments (LCAs) have been completed, examining a wide range of feedstocks, and to a lesser extent, conversion processes. LCA, ‘the compilation and evaluation of the inputs, outputs, and potential environmental impacts of a product system throughout its life cycle’ [1], has been applied to a wide variety of products and incorporated into transportation fuel regulations [2–4]. While considerable developments in lignocellulosic feedstocks and ethanol conversion process technologies have been made alongside concomitant advances in LCA methods and applications, considerable gaps remain in our understanding of the impacts of lignocellulosic ethanol production.

The aim of this review is to highlight key aspects of the lignocellulosic ethanol ‘pathway’, particularly differences in feedstocks, processes, co-products, and in LCA methods that could materially impact calculated GHG emissions. A case study for the production of ethanol from corn stover is used to illustrate the diverse GHG outcomes that can result from differing data, assumptions and analysis methods. The study incorporates specific literature data (from 2000 to present).

Life cycle assessment methods and their application to lignocellulosic ethanol

LCA was initially designed to examine environmental impacts of historical or current production over short, defined time periods to identify the largest impact reduction potential and improvement strategies without ‘burden shifting’. Traditional uses were to inform product development and policy. LCA is now also employed to enforce policy and to inform investors. LCA is the fundamental analysis tool to qualify fuels under a number of regulations (e.g., US Renewable Fuel Standard [2],...
European Union Renewable Energy Directive [3], California Low-Carbon Fuel Standard [4]). These initiatives mandate GHG emissions reductions for a regional fuel pool, or that an alternative fuel reduces GHG emissions by some prescribed level relative to an incumbent fuel.

While LCA approaches/methods have become more sophisticated since their inception, considerable methodological and application challenges remain [5,6]. There are two general types of LCA: attributional (aLCA) and consequential (cLCA). Both have a role in the analysis of lignocellulosic fuels, either via legislative initiatives or more broadly in the assessment of environmental performance. aLCA compares a candidate versus incumbent fuel based on physical relationships, while cLCA aims to determine overall impacts of the fuel’s use within a dynamic economic system using various economic approaches [7]. An attributional study might include consequential components (e.g., indirect land use change (iLUC) — see ‘Categories of lignocellulosic feedstocks’ below for more information) or include system expansion (rather than allocation) [8]. The US EPA RFS2 Regulatory Impact Analysis used the Forestry and Agriculture Sector Optimization Model (FASOM) and Farm and Agricultural Policy Research Institute (FAPRI) model to predict GHGs associated with land use change (LUC) from corn and cellulosic ethanol production, which were then added to base aLCA GHG emissions [2]. Consequential studies use emissions factors derived from aLCA studies to quantify emissions related to changes in the economic models underlying the cLCA approach. This is illustrated by Rajagopal and Plevin [9**, who investigated how adoption of renewable fuels impacted oil and agriculture markets. Martin et al. [10**] recently reviewed aLCA and cLCA studies of biofuels, focusing on system boundaries and noting distinctions among the models/studies. There is an ongoing debate about the relevance of each type of LCA to decision making [11**,12**,13,14].

LCAs of lignocellulosic biofuels in the literature are prospective, as the first facilities have only recently entered production, which creates challenges in applying LCA [5,6,15]. Major life cycle phases (e.g., biomass/ feedstock production, feedstock transportation, processing) and associated activities (e.g., on-farm fuel combustion) that have been included in many of the published LCAs of lignocellulosic ethanol from dedicated energy crops or agricultural residues are shown in Figure 1. LCAs have accounted for the emissions associated with these phases and associated cradle-to-gate (supply chain) activities (e.g., emissions associated with diesel fuel production for use in farm equipment). A key aspect of biofuels studies is the assumption that biogenic CO₂ emissions do not increase atmospheric CO₂ because the carbon released through lignin combustion, fermentation and ethanol combustion is exactly balanced with the carbon sequestered when the biomass regrows. Most lignocellulosic ethanol LCAs have focused on energy use and GHG emissions, and have compared ethanol use (in a low or high level blend with gasoline) in a light-duty vehicle to emissions of a reference gasoline vehicle. There are a few additional activities that are not shown in the figure that have been included in a small number of studies (e.g., transportation infrastructure).

Several reviews of lignocellulosic ethanol LCA studies have been completed [16–22]. Morales et al. reviewed over 100 LCA studies analyzing lignocellulosic ethanol, and concluded that the studies show a ‘clear reduction in GHG emissions.’ for ethanol compared to gasoline [23**]. The authors do caution that differences in methods chosen make comparing studies difficult. Borrión et al. [22] similarly concluded that results were dependent on system boundaries, functional units and allocation methods. Singh et al. [18] suggested that system boundaries, N₂O emissions, soil carbon dynamics and allocation methods for co-product credits are the greatest causes for uncertainty and variability.

**Categories of lignocellulosic feedstocks**

Lignocellulosic ethanol feedstocks are typically agricultural residues (e.g., stover, straw, bagasse), forestry products/residues (e.g., poplar, mill by-products), or dedicated energy crops (e.g., switchgrass, Miscanthus, energy cane). The production method and class of land for each category of feedstock affects overall GHG emissions [24]: some feedstocks are used directly, while others are residues of a primary crop (e.g., corn, wheat).

Agricultural and forestry residues are typically produced/removed during or after harvesting the primary ‘crop’. Concerns over what constitute sustainable residue harvesting rates arise due to soil erosion, reduced primary crop productivity, and soil nutrient and carbon depletion, have resulted in limits being placed on the fraction of residues that can be removed from fields, typically 25–70% for agricultural residues [25*,26,27]. Nutrient removal due to residue harvest can be countered through the addition of exogenous fertilizers. Agricultural and forestry residues may be treated as by-products of the main crop/tree cultivated and, as a result, emissions associated with feedstock production may be allocated to the primary crop/tree harvested rather than to the residual cellulose material. Upstream feedstock emissions, therefore, are normally restricted to those related to replacement nutrients and any additional energy required for residue harvesting (see Figure 1) [28]. Similarly, residues, which share cropland with the primary crop, are generally excluded from LUC impacts, with these impacts being allocated to the main marketable component of the material harvested. As a result, lignocellulosic ethanol produced from residues may have lower feedstock-related GHG emissions than that produced from dedicated energy feedstocks where the lignocellulosic material is the main marketable material.
Life cycle phases and associated GHG emissions typically included in life cycle inventory analyses of lignocellulosic ethanol from dedicated energy crops and agricultural residues in published literature.  

*Notes:* Major life cycle phases are indicated in blue. Dashed green boxes indicate biogenic carbon that is part of a closed cycle. ‘Carbon sequestered to soil’ may result in a net emission or sequestration depending on the specifics of the cases and feedstocks examined and is not part of a closed cycle. The processing stage (conversion of biomass feedstock to ethanol) assumes all electricity and heat requirements of the processing facility can be met through the use of the lignin and plant material that is not converted to ethanol. The sole co-product assumed is electricity. These processing assumptions are reflective of many lignocellulosic ethanol life cycle studies.

Adapted from Scown et al. [24].

However, in the future, as higher amounts of residues are used, it is possible that this approach will need to be re-examined, with energy and nutrient-related emissions assigned to each fraction of the plant (grain + residue). Residues may also eventually bear some of the burden attributed to LUC as their marketability grows.

Dedicated energy feedstocks often have higher yields than more traditional crops that are sources of residues, resulting in more efficient land use. Additionally, the root systems of dedicated energy crops are extensive and are left un-harvested, sequestering carbon and increasing soil carbon levels [21,29]. In contrast, sustainable use of agricultural residues is contingent upon establishing removal levels that do not deplete soil carbon. The magnitude of soil carbon losses (for residues) or gains (for dedicated energy crops) is just beginning to be quantified [17,30,31], but it is important to include in LCAs [24]. A concern related to large-scale production of biofuels from crops grown on land converted from other uses is LUC. Indirect LUC-related GHG emissions have been estimated in several LCA studies [32,33] but much uncertainty remains in accurately quantifying these emissions, for example, [34].

**The corn stover ethanol pathway: feedstock**

Corn stover is one of the most studied lignocellulosic feedstock options for ethanol production. Even so, pro-
cess-scale data for the production of corn stover and stover ethanol are relatively scarce. Few commercial scale lignocellulosic ethanol plants are in operation, and high-quality, reliable data needed for a detailed LCA are not publicly available.

To assess the current state of information for corn stover ethanol, literature data (from 2000 to present) were collected for production parameters that have the greatest impact upon corn stover ethanol (well-to-pump) GHG emissions, as predicted using the default pathway in GREET 2014 (see supporting information for methods and studies referenced). Our review assessed 80 peer-reviewed publications and government grey literature for the analysis. Parameter ranges from the literature are summarized in Table 1, indicating minimum, maximum, and average values. Literature data for corn stover harvest rates are common, while parameters such as farming energy for stover collection and N-fertilizer application are less frequently reported. Notably, there is an order of magnitude difference between the minimum and maximum values for stover harvest rate, the maximum value for farming energy for stover collection is more than four times the minimum value and there is a two-fold difference in projected N-fertilizer use, the latter is likely associated with the assessed N content of the corn stover, which also varies widely.

Corn stover is predominately handled as a residue from corn grain production, and as a result, a significant portion of upstream agricultural inputs and emissions are not attributed to the stover. Farming energy for stover production has largely been restricted to the energy required to harvest stover from the field, while inputs associated with the cultivation and harvesting of corn are allocated to corn grain [35]. Similarly, N-fertilizer emissions for corn stover are generally restricted to the amount of fertilizer needed to replace the nitrogen removed from the field by stover harvest [15,35]. As a result, corn stover carries fewer energy and emissions burdens for upstream processes than many other cellulosic feedstocks. Recent studies [28,31,36,37,38] have examined allocation of a portion of corn production-related emissions to stover, and found this can have considerable impacts on the resulting ethanol GHG emissions.

### Lignocellulosic ethanol conversion/production processes

The process technology has several direct and indirect impacts on GHG emissions and other metrics typically evaluated via LCA. However, most LCAs are narrowly based, incorporating the US Department of Energy’s National Renewable Energy Laboratory’s (NREL) process that uses dilute acid pretreatment, with electricity as the sole co-product [39]. Nonetheless, several technology options have been proposed and adopted for the commercial plants, with additional technologies considered at the pilot scale [40]. On the surface, these differ primarily in the area of pretreatment, but there are cascading impacts on downstream processes, including enzymatic hydrolysis and fermentation, and ultimately, the yield of ethanol and the yield (and type) of co-products.

### Biomass pretreatment

There is no consensus best practice for biomass pretreatment. The original NREL process, and several pilot and commercial operations since then, have adopted dilute acid pretreatment technology, generally by pre-soaking the biomass in dilute sulfuric acid, followed by steam explosion. Others have adopted either an alkaline pretreatment or an autohydrolysis process that relies on either steam or hot water for pretreatment. Using vapor phase versus liquid phase pretreatment affects water and energy use, and downstream solids loading [41]. Vapor

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### Table 1

<table>
<thead>
<tr>
<th>Parameter b</th>
<th>Unit</th>
<th>Number of data points</th>
<th>Parameter value</th>
</tr>
</thead>
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<tr>
<td>Electricity co-product</td>
<td>kWh/L EtOH</td>
<td>21</td>
<td>0.0 1.03 0.53</td>
</tr>
<tr>
<td>N content of stover c</td>
<td>kg/dry tonne feedstock</td>
<td>7</td>
<td>5.47 8.26 6.7</td>
</tr>
<tr>
<td>N-fertilizer use</td>
<td>kg/dry tonne feedstock</td>
<td>7</td>
<td>4.96 12.12 7.56</td>
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<tr>
<td>NaOH consumption</td>
<td>kg/dry tonne feedstock</td>
<td>2</td>
<td>0.0 0.03 0.02</td>
</tr>
<tr>
<td>Cellulase consumption</td>
<td>kg/dry tonne feedstock</td>
<td>9</td>
<td>1.24 37.4 6.72</td>
</tr>
<tr>
<td>Farming energy for stover collection</td>
<td>MJ/dry tonne feedstock</td>
<td>8</td>
<td>21.4 897 487</td>
</tr>
<tr>
<td>Corn steep liquor</td>
<td>kg/dry tonne feedstock</td>
<td>9</td>
<td>1.7 17.3 15.3</td>
</tr>
<tr>
<td>Stover harvest rate</td>
<td>Dry tonne/ha</td>
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<tr>
<td>N-N₂O conversion</td>
<td>% of N</td>
<td>7</td>
<td>0 2.34 1.21</td>
</tr>
<tr>
<td>Ethanol yield</td>
<td>L/dry tonne</td>
<td>43</td>
<td>137 449 329</td>
</tr>
</tbody>
</table>

a Details on methods and literature sources utilized to develop the table are in the supporting information.
b Parameters listed in order of sensitivity/impact upon well-to-pump GHG emissions calculated via the default GREET 2014 model pathway with MRO electricity region selected.
c Feedstock-related parameters are highlighted in grey.
phase pretreatment processes tend to be more energy efficient due to improved downstream heat recovery [41]. The type and conditions of pretreatment also affect cellulose activation (which affects enzyme use), and the degree of hemicellulose converted to inhibitors (affecting yield) [40]. Pretreatment processes that use acids or bases as catalysts require a subsequent neutralization step before enzymatic hydrolysis, increasing waste product load [41]. Recovery and reuse of the catalyst may be needed for financial reasons, but increases process energy requirements. A key advantage of the steam explosion process (without added chemicals) is that the high pressure steam added during pretreatment can be recovered and used as an energy source downstream, for example, for distillation. Liquid pretreatment processes or vapor processes using acids or bases that require neutralization or recovery are less likely to be able to employ such heat integration.

Enzyme hydrolysis
Key aspects of enzyme hydrolysis that affect GHG emissions and energy intensity include the solids loading, and the amount of enzyme and energy required for sufficient mixing. In Table 1, well-to-pump GHG emissions are noted to be sensitive to cellulase (enzyme) consumption, and a wide range of values is reported in literature (1.24–37.4 kg cellulase/dry tonne feedstock). Higher solids loadings are more difficult to mix, and require more enzymes (IMN Gaona et al., unpublished), but reduce the amount of energy needed for pumping, downstream fermentation and distillation. Process designs that allow rapid viscosity reduction of a high solids biomass slurry using the smallest possible quantity of enzymes and low rpm agitation will have a significant advantage increasing the amount of excess electricity available for export. The amount of enzyme used has a direct effect on GHG emissions due to the carbon intensity of the enzyme production process [42,43]. Hydrolysis conditions also dictate the conversion of glucose and xylose into fermentable sugars, and consequently, the ethanol yield. Since most LCA metrics are calculated per unit ethanol (energy or volume), process yields are important.

Fermentation
Most technologies now use advanced fermentation organisms to convert glucose and xylose into ethanol, albeit incompletely, and occasionally, via separate fermentation trains [15,39,44]. Although separate fermentation trains tend to be less energy efficient, they may be required to ensure suitable ethanol yields. Pretreatment and hydrolysis processes that preserve hemicellulose and increase xylose production also generate higher yields and ethanol titers that require less energy for distillation.

Distillation
Most processes employ conventional distillation methods. However, choices upstream regarding the process hydraulic load, pretreatment, enzyme hydrolysis, and fermentation can materially impact the energy intensity of the ethanol purification process, and thus the amount of electricity that can be generated as a co-product [44].

Co-products
Most operations are predicated upon on-site energy generation using residual lignin (which also includes unconverted cellulose, hemicellulose and other carbohydrates). If the ‘lignin’ satisfies process demands for steam (or hot water) and electricity, the excess electricity may be sold to the grid as a co-product (see Figure 1). In most cases, the co-product electricity is assumed to displace high carbon intensity electricity on the US Midwest grid. Thus, the ethanol yield and the energy intensity of the process, both of which affect the amount of electricity available as a co-product, can have a significant effect on the GHG intensity of the lignocellulosic ethanol process [44]. This is reflected in the electricity co-product being the parameter in Table 1 that most impacts well-to-pump GHG emissions and the wide variation in values for this parameter.

Decisions to produce alternate co-products, such as pellets, xylitol, or animal feed/proteins, can also materially impact the GHG intensity and energy intensity of the process, depending in large part upon the carbon intensity of the products that are displaced. GHG reductions for ethanol of 60–140% relative to gasoline have been noted, depending upon the conversion process and co-product selection [44]. Co-product treatment (through allocation, system expansion, etc.) is a key methodological choice that affects the outcome of LCA studies [5], and plays a major role in lignocellulosic biofuels’ life cycle GHG emissions [44,45].

The corn stover ethanol pathway: conversion process
Despite the numerous literature sources that discuss corn stover ethanol, independently generated data for process chemical usage (enzymes, corn steep liquor, etc.) are comparatively scarce (Table 1). Three of the top five parameters most impacting well-to-pump GHG emissions are associated with the conversion process. Many sources present process data from NREL models and reports on the production of lignocellulosic ethanol from corn stover, using dilute acid pretreatment [39,46,47]. While the NREL models are comprehensive, they are theoretical and have not been validated at an industrial scale. This is a concern because many of these chemicals, including enzymes, have high GHG intensities, and their impacts on life cycle emissions may not be accurately quantified [42,43].

Indirect effects of lignocellulosic ethanol
Consequential LCA has brought to the forefront attention to the inclusion of indirect effects in LCA. Potential
indirect or market-mediated effects of lignocellulosic ethanol production are not limited to iLUC. Other possible market-mediated consequences include the effect of increased ethanol production on gasoline consumption and price [48] and land prices [24]. These effects have received much less attention in the literature than iLUC. Regarding fuel market effects, LCAs comparing GHG emissions of ethanol to those of gasoline typically assume displacement on an energy equivalent basis (i.e., 1 MJ ethanol replaces 1 MJ gasoline). However, if an increase in ethanol consumption causes gasoline prices to decrease (the market response), it may cause an increase in consumption of fuel, including gasoline [48]. This can occur locally, if the biofuel has a lower price per energy equivalent unit than gasoline, or internationally if the mandated biofuel is priced higher than gasoline on an energy basis. In either case, the supply of fuel is increased (even temporarily), reducing fuel prices and increasing demand. When this change is accounted for, 1 MJ of ethanol may displace less than 1 MJ of gasoline. The fuel market impact is independent of the feedstock used for ethanol production, and applies to fuels produced from cane, lignocellulosic or corn (grain) crops. With time, this impact can decrease with a reduction in gasoline production or a general increase in fuel use, for example, more drivers as the population increases.

Conclusions and perspectives

Advances in lignocellulosic ethanol feedstocks and technology, including commercialization, have coincided with increased attention to environmental impacts. Use of LCA to assess GHG emissions reductions has played an increasingly prominent role in various policies, including the European Renewable Energy Directive, the US Renewable Fuels Standard, and California’s Low-Carbon Fuel Standard. In spite of this progress, there remain critical unresolved issues identified within this review that are expected to impact the future of lignocellulosic ethanol and its energy/GHG emissions performance. Important questions have arisen regarding feedstock-related emissions, consequential versus attributional life cycle aspects, choice of system boundaries, and allocation methods. Decisions regarding feedstock, process technology and co-products can materially impact GHG emissions calculations. The carbon intensity of the product displaced by the co-product also plays an important role if employing system expansion methods.

Considering the case study of corn stover ethanol with lignin-derived electricity as a co-product, predicted life cycle (well-to-pump) GHG emissions vary widely due to large variability/uncertainty in a number of key parameters, including the amount of electricity produced, N-related emissions due to supplemental fertilizer requirements and the N content of stover, cellulase requirements, farming energy, and ethanol yield, among others.

The future holds promise as lignocellulosic ethanol facilities, and their associated supply chains, are now a reality in several countries, allowing for the collection of more definitive feedstock, transportation and production data for LCA. In addition, ongoing method development/refinement in LCA (both for aLCA and cLCA) will facilitate more comprehensive/accurate studies, and importantly, those emphasizing transparency and explicitly characterizing uncertainty and variability.

Acknowledgements

BAS and HLM acknowledge financial support from the Natural Science and Engineering Research Council of Canada and BioFuelNet Network Centre of Excellence. WMG is supported by the Center for Climate and Energy Decision Making (CEDM), through a cooperative agreement between the National Science Foundation and Carnegie Mellon University (SES-0949710).

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.copbio.2015.12.021.

References and recommended reading

Papers of particular interest, published within the period of review, have been highlighted as:

- _of special interest_
- _of outstanding interest_


The paper challenges the assumption of a one MJ per MJ replacement of gasoline for ethanol. The research shows that displacement of gasoline use locally via mandate or via carbon intensity based regulations resulted in increased fuel use in the rest of the world and an overall increase in petroleum use. Biofuels mandates increased global greenhouse gas emissions and carbon intensity standards generally decreased emissions.

The authors review recent LCA studies for the production of biofuels from various energy crops and wastes. They compare attributional results and consequential results, and discuss the impacts of system boundaries and other important LCA modeling decisions. There is a very good summary of the current debate regarding these methods and implications for biofuels analysis.


The authors compare attributional and consequential LCA approaches, suggesting that cLCA is a conceptually superior method for estimating actual changes in the real world compared to eLCA, while recognizing that there are limitations to cLCA results. They make general recommendations for analyzing and interpreting LCA results for policy makers to make sound environment-related decisions.


The paper challenges several points in Plevin et al. [11], which claims cLCA is the superior methodology that closely reflects real-life LCA. The authors suggest that LCA modeling should not be limited to two approaches that exclude studies that do not fall into either of the categories. The notion of incorrect models and conceptual superiority of an approach limit the development of LCA frameworks that better fit the biofuel system.


A review of available lignocellulosic ethanol LCAs to compare ethanol’s environmental impacts relative to conventional fossil fuels as well as first generation bioethanol. Generally, studies showed a clear reduction in GHG emissions with a positive energy balance for the lignocellulosic ethanol. The authors also pointed out the difficulty in comparing LCA results due to variations in underlying methodologies.


25. Graham RL, Nelson R, Sheehan J, Perlack RD, Wright LL: Current and potential U.S. corn stover supplies. Agron J 2007, 99:1. The authors examine effects of corn stover collection on erosion, crop productivity, and soil carbon and nutrients. Location and quantity of corn stover that can be harvested sustainably including collection costs are estimated. The authors report that changes in land management could increase the amount of sustainable supply of corn stover and provide sufficient feedstock for a biorefinery in the Midwest U.S.


34. Plevin RJ, O’Hare M, Jones AD, Tom MS, Gibbs HK: Greenhouse gas emissions from biofuels’ indirect land use change are uncertain but may be much greater than previously estimated [Internet]. Environ Sci Technol 2010, 44:8015-8021.


The paper examines the effects of spatial variation of biomass supply, process input supply chains, and ongoing technology development on the life cycle greenhouse gas emission of corn stover ethanol. A comparative analysis of the NREL 2002 and 2011 conversion process models is also presented, highlighting the key differences that affect the emissions results.


A review of updated enzyme production details and their inclusion in the GREET model. The authors emphasize the importance of including enzymes and yeast in biofuel LCAs.


