Life-Cycle Assessment of Bioethanol from Pine Residues via Indirect Biomass Gasification to Mixed Alcohols*

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Abstract

The goal of this study was to estimate the greenhouse gas (GHG) emissions and fossil energy requirements from the production and use (cradle-to-grave) of bioethanol produced from the indirect gasification thermochemical conversion of loblolly pine (Pinus taeda) residues. Additional impact categories (acidification and eutrophication) were also analyzed. Of the life-cycle stages, the thermochemical fuel production and biomass growth stages resulted in the greatest environmental impact for the bioethanol product life cycle. The GHG emissions from fuel transportation and process chemicals used in the thermochemical conversion process were minor (less than 1 percent of conversion emissions). The net GHG emissions over the bioethanol life cycle, cradle-to-grave, was 74 percent less than gasoline of an equal energy content, meeting the 60 percent minimum reduction requirement of the Renewable Fuels Standard to qualify as an advanced (second generation) biofuel. Also, bioethanol had a 72 percent lower acidification impact and a 59 percent lower eutrophication impact relative to gasoline. The fossil fuel usage for bioethanol was 96 percent less than gasoline, mainly because crude oil is used as the primary feedstock for gasoline production. The total GHG emissions for the bioethanol life cycle analyzed in this study were determined to be similar to the comparable scenario from the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation model. A sensitivity analysis determined that mass allocation of forest establishment burdens to the residues was not significant for GHG emissions but had significant effects on the acidification and eutrophication impact categories.

The United States is the largest bioethanol producing country, with 49.2 billion liters as of 2010 (Renewable Fuels Association [RFA] 2010). Controversy around conventional biofuels, which are usually produced through the conversion of corn grain (USA) and sugar cane (Brazil) (Mitchell et al. 2008, Gonzalez et al. 2011), has prompted research and investment in advanced biofuels produced from non-foodbased feedstocks, including lignocellulosic material. Additionally, lignocellulosic biomass is an important feedstock for other types of bioenergy (wood pellet, briquettes,

biopower). Studies suggest that the use of lignocellulosic feedstocks (e.g., agriculture and urban-derived residues and forest feedstocks) has clear benefits in the mitigation of greenhouse gas (GHG) emissions (Schneider and McCarl 2003, Zhang et al. 2009). Greenhouse gases, primarily made up of CO_2 , N₂O, CH₄, H₂O vapor, and O_3 , are thought to be the active catalyst in the documented rising global temperatures (Oliver et al. 2009, Solomon et al. 2009).

The US Energy Independence and Security Act (EISA 2007) outlined a set of goals to increase energy security and

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reduce GHG emissions. Biofuels from lignocellulosic material have the potential to be an important component of the solution to reducing fossil fuel consumption and GHG emissions (Hahn-Hagerdal et al. 2006, Sims et al. 2010). For this reason, the Renewable Fuels Standard was passed into law in the United States in 2005 and requires commercial production of approximately 136 billion liters of blended renewable transportation fuels by the year 2022. However, there remains controversy surrounding the production of first-generation biofuels and potential GHG savings as the production and use of conventional biofuels (e.g., corn grain derived) can have a negative net energy ratio, meaning that more fossil fuel energy may be consumed to produce and transport conventional biofuels than the energy produced (Davis et al. 2009).

The EISA also requires the US Environmental Protection Agency to create and enforce life-cycle GHG threshold standards to ensure reductions through the use of renewable fuels. By achieving these threshold requirements (60% GHG reduction for cellulosic bioethanol and 20% for other biofuels, such as cornstarch-derived bioethanol), the displacement of imported fossil fuel with domestic biofuel will decrease anthropogenic GHG emissions and promote more sustainable development in the energy sector of the US economy.

In 2001 the Argonne National Laboratory developed the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model to analyze the GHG emissions from the biofuels life cycle, using many different production pathways and blending options (Wang 2001). GREET was specifically designed for transportation emissions calculations; however, it also incorporates life-cycle concepts and data from raw material extraction and manufacturing upstream emissions prior to the use phase. GREET has been the backbone for a large part of the existing knowledge surrounding biofuel GHG emissions. Data created with the GREET model have added significantly to our understanding of biofuel GHG emissions but do not encapsulate all biofuel conversion routes and only allow for the model user to incorporate limited conversion facility data. Additional incorporation of specific facility operational data may be required to attain an accurate depiction of GHG emissions for that facility.

Despite the nonspecific nature of GREET model output data, the GREET model serves as an excellent platform for comparing GHG emissions from a select set of common production processes. For emerging technologies such as biomass gasification and synthesis of syngas, the GREET model and other similar tools become less useful because generic process emissions data will no longer apply to more optimized or unique unit processes or conversion technologies. More discerning and robust methodologies and database values need to be developed to address these currently un-commercialized technologies.

Life-cycle assessment (LCA) is the primary tool used to analyze the GHG emissions from transportation fuels, such as biofuel or gasoline. The GHG emissions values for petroleum fuels are well established (Zhou et al. 2007, Nanaki and Koroneos 2012); however, there is not yet consensus in the literature surrounding emissions from emerging biomass conversion technologies, in part because of rapid innovation and the lack of existing large-scale commercial conversion facilities or common facility designs (Banerjee et al. 2010, Gibbons and Hughes 2011).

Forest residues, a waste product from forest logging and forest management activities, are available in the United States at an annual rate of 62 million metric dry tonnes (Perlack et al. 2005, Perlack and Stokes 2011). Forest residue is often described as biogenic, or carbon neutral, meaning that the carbon emissions associated with the direct burning of the biomass or products derived from the biomass are not considered GHG emissions. This disputed claim of carbon neutrality often found in literature associated with lignocellulosic bioethanol and other forest products has been recently questioned as a result of studies exploring the impact of land use change (LUC) and biogeochemical emissions from forest harvest and biomass removal (Fargione et al. 2008; Searchinger et al. 2008, 2009). This study, however, does not allocate LUC or harvest burdens to the forest residues or bioethanol production life cycle because the residues are herein considered a waste stream. Thus, combustion emissions are also considered biogenic $CO₂$ equivalent emissions and are offset with carbon captured during residue growth. This assumption is justified because leaving forest residues after harvest, the alternative to collection of forest residues and biofuel production, releases atmospheric $CO₂$ during natural decomposition on the forest floor (Sullivan et al. 2008).

From the 62 million metric dry tonnes, Perlack and Stokes (2011) estimated an annual availability of 39 million metric dry tonnes at less than \$91 per metric dry tonne (\$100 per dry short ton) delivered, the indicated threshold for feedstock delivery feasibility. Feedstock delivered cost and supply chain are currently being further explored for forest residues and other feedstock types by the present authors (Daystar et al., submitted for publication). This biomass stream, in addition to others such as roundwood, municipal solid waste, recycled wood, and paper, could supply the biomass required to feed the emerging bioenergy industry (Miao et al. 2012, You et al. 2012). The study herein analyzed a base case of processing forest residues from pine plantations only into bioethanol; however, for continual operation of a scaled-up facility, the residue availability, transportation distance, cost, and environmental burdens would likely necessitate a mixture of feedstocks to be converted as part of the facility's standard operating procedures. This, in turn, would necessitate a flexible conversion process (Jameel et al. 2010).

The goal of the study herein was to quantify the environmental burdens of bioethanol produced from gasification of forest residues on a cradle-to-grave basis. The findings from this study will enable policy makers, stakeholders, and the emerging biofuels industry to make informed decisions surrounding the environmental impacts of large-scale biofuel production and use within the Southeastern United States.

Methods

Goal and scope

The goal of this study was to examine select environmental impacts resulting from the production and use of bioethanol manufactured from forest residues using a thermochemical conversion process on a cradle-to-grave basis. The environmental impacts, calculated as GHG emissions (kilograms of $CO₂$ equivalents per megajoule), eutrophication (N equivalents per megajoule), acidification $(H +$ equivalents per megajoule), and fossil fuel–based

Figure 1.—Cradle-to-grave system boundaries (note: "T" represents transportation processes). The dashed line encompasses the unit processes evaluated, while the unit processes outside the dashed line are associated with other forest products not considered within the scope of this study.

energy usage (megajoules of fossil fuel input per megajoule of fuel produced), were compared with those of gasoline. This study also compared GHG emissions reductions with the required reductions outlined in the Energy Independence and Security Act (EISA 2007) to predict the feasibility of using forest residues as part of the US biofuels portfolio. The Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI; Bare et al. 2003, Jolliet et al. 2004) impact assessment method was used within the SimaPro 7.2 calculation framework to quantify environmental impacts (Pré Consultants 2010).

To compensate for differing fuel heating values, a functional unit of 1 MJ (higher heating value) of combusted transportation fuel was selected. An energy-based functional unit ensured an equal comparison by removing influences of energy density between fuel types (Oak Ridge National Laboratory [ORNL] 2012).

System boundaries

In order to quantify the overall GHG emissions from the production and use of bioethanol produced via gasification of forest residues, a cradle-to-grave system boundary was selected. The unit processes evaluated within this study include carbon absorption during tree growth, residue collection after pine harvest, transportation of the biomass to a conversion facility, thermochemical conversion processes, transportation to end user, and combustion (Fig. 1). The system boundary shown in Figure 1 was chosen in methodological agreement with previous LCA studies of biofuel production systems to exclude forest management prior to collection and to incorporate only first-tier upstream burdens (Neupane et al. 2011, González-García et al. 2012). Thus, some portion of the burdens associated with the manufacture of the motor that transports the chipped forest residues within the conversion facility was not allocated to the production of the bioethanol product, as an example. System expansion was used where possible, and allocation was used for a sensitivity analysis of the impact of system expansion to net emissions only.

.Avoiding allocation of possibly unfair credits or burdens, the biomass thermochemical conversion process was simulated to neither consume electricity nor return electricity to the US electrical grid as a result of coproduction of syngas or char from the gasification

process. Previous techno-economic studies, both modeled and bench scale, have shown that co-combustion of biomass or produced syngas can equalize the energy demand and supply within the biomass conversion facility to make the facility energy self-sufficient, requiring no additional fossil fuel–based energy inputs (Kumar 2009, Dutta et al. 2012). This assumes that char from the gasification process and a percentage of uncleaned syngas are combusted for combined heat and power on site (Whitty et al. 2008, Seiler et al. 2010, Jett 2011). In this study, the amount of syngas and char sent to the on-site boiler was scaled to accommodate the onsite steam and electricity needs. In this way allocation issues surrounding grid electricity, coal, and natural gas energy use or creation during the conversion process were avoided.

Life-cycle impact assessment method

Process simulation models in Aspen Plus (see below) were used to generate inventory results for the thermochemical conversion process used in the life-cycle impact assessment (Phillips et al. 2007). The TRACI impact assessment method was used because it is a US-specific impact assessment method (Bare et al. 2003). SimaPro was used to calculate the final impacts using US Life-Cycle Inventory (USLCI) data and the TRACI impact assessment method (Pré Consultants 2010). SimaPro is an LCA calculation software program that uses the USLCI of emissions and impact data from the US Department of Energy (US DOE) National Renewable Energy Laboratory (NREL) and uses the TRACI impact assessment method to interpret the quantified impacts.

Life-cycle stages description and study assumptions

Data quality.—Because no commercial biomass gasification to mixed alcohols conversion facilities currently exist, process simulations, reports, studies, and the USLCI database provided the necessary data. Processes and emission factors specific to the United States were used, with the exception of magnesium oxide emissions, which were taken from European data.

Study assumptions.—The following assumptions were made in this study.

- Pine plantations are sustainably managed, meaning no change in productivity from year to year, and no land use changes.
- * Forest residues are considered to be a waste stream of timber/pulpwood production.
- Pine forest establishment, maintenance, and harvest environmental burdens were allocated completely to the timber/pulp products for the base case.
- Belowground carbon and biomass is considered to be at steady state with no biogeochemical carbon loss as a result of use of forest residues, since residues are considered a waste stream.
- Avoided residue decomposition emissions from alternative use of residues are not allocated as a credit to the bioethanol life cycle.
- Higher heating value of gasoline was assumed to be 35 MJ/liter (43.8 MJ/kg; ORNL 2012).
- Higher heating value of bioethanol was assumed to be 24 MJ/liter (29.8 MJ/kg; ORNL 2012).

Feedstock production.—Forest residues consist of the tops, small branches, and leaves of the harvested trees deemed unusable for pulpwood or saw timber. This unmerchantable material is collected from the forest at the time of harvest using timber harvesting roadways. Depending on the forest characteristics and machinery technology, a removal rate of the total residues of 50 to 65 percent is expected (Perlack and Stokes 2005). Plant material including leaves and other smaller material are left in the forest to decompose, partially into soil carbon and other nutrients required for tree growth and partially as biogenic atmospheric $CO₂$.

Loblolly pine (*Pinus taeda*) plantations grown in the Southeastern United States are primarily intensively managed and privately owned (Andreu et al. 2011). Forest operations for these intensively managed plantations typically include site preparation, seedling planting, fertilization, and herbicide applications (Jokela et al. 2010). Another common technique includes forest thinning: removing a portion of the biomass in the middle of the production cycle (Jokela et al. 2010, Andreu et al. 2011). These forest operations, along with timber harvesting, will produce GHG emissions prior to collection of the forest residues. For this study, GHG emissions from forest operations (to grow and harvest the wood) were 100 percent allocated to the main products (timber or pulp logs). The forest residues were assumed to be a waste stream; therefore, allocation of the LUC, establishment, maintenance, and harvest GHG emissions to the biofuel production from the forest residues process were not included for the base-case scenario.

Several recent studies have suggested that LUC, establishment, and maintenance activities should be considered when calculating life-cycle burdens due to nutrient and carbon flux in soil after removal of residue (Repo et al. 2011); however, these findings are heavily dependent upon the assumption of a relatively slow forest residue decomposition rate for which consensus has not been reached (Eriksson et al. 2007, Wall 2008, Luiro et al. 2010). On the contrary, other studies have found decomposition rates to be fairly rapid (Wang et al. 2002, Palviainen et al. 2004, Sathre and Gustavsson 2011), which would support the assumption that emissions from precollection activities should be taken into account. Owing to this uncertainty, a sensitivity analysis was conducted that allocates precollection emissions to the bioethanol life cycle based on mass ratios.

Although activities associated with the management of intensive pine plantations emit GHGs, $CO₂$ is captured from the atmosphere and stored as wood and plant material during tree growth. Therefore, carbon dioxide captured within the biomass is accounted for within this study as a negative emission, offsetting GHG emissions from other life-cycle stages. This technique has been used in previous studies with broad acceptance when used consistently (Lemus and Lal 2005, Farrell et al. 2006, Tilman et al. 2006, Liebig et al. 2008). To calculate the carbon dioxide stored in the biomass during growth, the forest residues were estimated from literature to be 50 percent carbon (Kilpelainen et al. 2011). Using molecular weight ratios, the $CO₂$ absorption was calculated per tonne and included as a negative emission in the net GHG emissions calculation.

Feedstock collection.—Emissions from feedstock production and harvesting originate from fuels and lubricants used by equipment required for residue collection and chipping. The equipment used during collection included skidder, feller/buncher, and chipper. Equipment manufacturing emissions were determined to be outside the scope of the study and were not included. The USLCI record name used for these calculations was ''whole tree biomass chipping'' submitted by the Consortium for Research on Renewable Industrial Materials (CORRIM; Johnson et al. 2012).

Feedstock transportation.—Transportation distances were assumed to be 80.4 km (50 mi) from forest to facility. The actual transportation distance will depend on biomass availability and facility processing rates. An assumption of 80.4 km (50 mi) for 700,350 metric dry tonne eq per y was calculated using the methods described in Gonzalez et al. (2012). Emissions from empty trucks returning from the facility gate to the point of collection were assumed using a separate emissions value for the empty truck transport and the same transportation distance of 80.4 km. More detailed biomass feedstock production model studies from the authors have recently been published (Gonzalez 2011, Gonzalez et al. 2012) and are in manuscript (Daystar et al., submitted for publication). GHG, eutrophication, and acidification emission factors for a diesel combination truck were taken from the USLCI database (NREL 2003).

Thermochemical conversion process and Aspen model.— The NREL thermochemical bioethanol production process (Phillips et al. 2007) was the conversion pathway used for this analysis and simulated in Aspen Plus. Aspen Plus is a chemical engineering and energy production process simulation software program that enabled the authors to explore the constraints and parameters of an industrialscaled gasification process for specific biomass feedstock inputs and operational parameters (AspenTech 2012). Simulation modifications were required to meet the unique needs of this study as described in Gonzalez et al. (2012). The continuous feedstock supply was set at 700,350 metric dry tonne equivalents of forest residue biomass per year (772,000 dry short tons per y), and a moisture content of 45 percent was assumed from the literature (Jameel et al. 2010, Patterson et al. 2011). Both ultimate and proximate analyses of the feedstock were required to run the model (Table 1). Compositions of loblolly pine (US DOE 2005, Jameel et al.

Table 1.-Loblolly and hybrid poplar ultimate and proximate analysis compositions.^a

| | Ultimate analysis (wt%, dry basis) | | | | Proximate analysis (wt%, dry basis) | | | Moisture content | | |
|-----------------------------------|------------------------------------|-----|-----|----|-------------------------------------|-----|----------------|-------------------|-------|--------------------|
| Feedstock type | | | | | | Ash | % fixed carbon | % volatile matter | % ash | $(\%$, wet basis) |
| Hybrid poplar (NREL) ^b | 51 | 6.0 | 0.2 | 42 | | 0.9 | | 84 | 0.87 | 45 |
| Loblolly forest residues | 52 | 6.5 | 0.0 | 41 | 0.0 | 0.4 | | 85 | 0.40 | 45 |

^a Sources: US Department of Energy (2005) and Phillips et al. (2007).

 b NREL = National Renewable Energy Laboratory.

2010) and hybrid poplar (the NREL base-case feedstock shown for general reference) are listed in Table 1. Residues were considered to have similar composition as compared with the rest of the tree, although a sensitivity analysis of this assumption should be conducted in future work, using pine residue composition data taken from the literature (Frederick et al. 2008, Kilpelainen et al. 2011).

The thermochemical process is separated into seven major process areas within the Aspen Plus simulation (Fig. 2). Each process area is composed of multiple unit processes, such as reactors, separations, heat exchangers, and other operations that alter the matter within the process. These major process areas are briefly described below; however, a detailed description can be found in Phillips et al. (2007).

The biomass delivered to the conversion facility is first dried in the feedstock handling and drying stage to a moisture content of approximately 5 percent. The dried, chipped biomass is then reacted with steam and extreme heat from contact with heated olivine sand in the gasification process to produce synthesis gas, mainly consisting of CO and $H₂$. The syngas is then removed from the char and steam using a cyclone, which separates the char and sand from rising syngas. Clean syngas is then sent to the alcohol synthesis process to react with methanol to form ethanol and higher alcohols. Some additional catalyst fouling compounds and tar are formed in this process. These compounds are removed

through the gas cleanup and conditioning process to produce smaller carbon-chain molecules and co-combusted to heat the olivine sand. A full list of processing parameters and a detailed process description is included in Phillips (2007) and Jameel et al. (2010).

Fuel transportation.—CO₂ emissions from diesel-powered combination truck (tractor trailer) transportation were calculated using the USLCI database emission factors. Total transportation distance was assumed to be 80.4 km one way from fuel production plant to fuel pump.

Fuel combustion.—Fuel use emissions data were based on combustion of the produced bioethanol in a light-duty passenger vehicle, and data were obtained using the GREET model (Wang 2001). Emissions from combustion of pure bioethanol fuel were calculated; however, perfectly pure bioethanol is not currently practical because of incompatibility with available infrastructure and vehicles. The pure bioethanol comparison with gasoline shows the maximum GHG savings possible from biofuel production and use. These emission savings could be used to classify biofuel as an ''advanced biofuel'' under EISA biofuel classifications (EISA 2007). GREET emissions data were generated using primarily default values for study-specific feedstock and flex fuel vehicle information. For example, emissions data specific to ethanol combustion were used for the 100 percent bioethanol scenario (no blending). The gasoline

Figure 2.—Thermochemical bioethanol production process flow diagram based on the National Renewable Energy Laboratory model.

Table 2.—Greenhouse gas (GHG) emission factors for process chemicals and nonwood inputs.

| Material/process | Unit | Emission factor | kg $CO2/MJ$ bioethanol (HHV) ^a |
|--|------------------------|-----------------|---|
| Magnesium oxide ^b | kg CO ₂ /kg | 3.77 | $1.42E - 05$ |
| Olivine ^c | kg CO ₂ /kg | $3.92E - 02$ | $2.34E - 05$ |
| Molybdenum ^c | kg CO ₂ /kg | 0.108 | $1.30E - 05$ |
| Waste treatment ^c | kg CO ₂ /kg | $6.37E - 07$ | $8.77E - 10$ |
| Landfill transportation ^c | kg $CO2/30$ km | 0.129 | $6.93E - 06$ |
| Landfill ^c | kg CO ₂ /kg | $2.45E - 03$ | $2.19E - 07$ |
| Total chemical and waste GHG emissions | | | $5.78E - 05$ |
| % of fuel production GHG emissions | | | 0.02 |

^a HHV = higher heating value.
^b Sources: World Resources Institute and World Business Council for Sustainable Development (2004) and Integrated Pollution Prevention and Control (2010).

^c Source: National Renewable Energy Laboratory (2003).

equivalent for comparison was assumed to be 50 percent reformulated and 50 percent conventional fuel.

Results

Life-cycle inventory

Process simulation.—Using the thermochemical conversion simulation, the material and energy balances of the process were calculated with a 98.5 percent closure. This means that measured input energy and matter very nearly equaled measured output energy and matter and indicates that efficiency of conversion was not skewed as a result of calculation errors, theoretical energy, mass loss, or creation. Alcohol yields from the simulation of loblolly pine residue conversion revealed that 369 liters of ethanol and 65 liters of propanol are expected alcohol yields per ovendry metric tonne of biomass converted. Both ethanol and propanol are shown because of the high percentage of both in the mixed alcohol produced. The propanol, however, was converted to ethanol equivalents using a multiplier based on the higher heating value ratio of these two alcohols. The total yield in ethanol equivalents was projected to be 424 liters per bone dry (BD) metric tonne of biomass converted. Spatari et al. (2010) estimated a range of 250 to 350 liters/BD tonne, and Dutta et al. (2012) calculated 355 liters/BD tonne through indirect gasification and mixed alcohol synthesis, which was the conversion technology modeled here.

Emissions from production and treatment of chemicals and waste used during the thermochemical conversion process (gasification) were calculated and determined to be not important to net environmental burdens across the life cycle of the bioethanol product. Both emission factors and overall emissions from each chemical are listed in Table 2. All GHG emissions from process chemicals and waste treatment represented only a small fraction of the overall emissions. These values are consistent with previous studies (Bright and Strømman 2009, Reijnders and Huibregts 2009, Puy et al. 2010, Whittaker et al. 2011, Gonzalez et al. 2012). These studies determined that cultivation and harvest, conversion, and combustion (for studies not considering biogenic $CO₂$ separately from anthropogenic $CO₂$) were the largest contributors to total GHG emissions across the life cycle.

Biomass productivity.—Biomass productivity in terms of ovendry (OD) equivalent tonnes per hectare and bioethanol yield per hectare were calculated (Table 3). Data from loblolly pine growth in the Southeast (Allen et al. 2005) were used as a basis for tonnes per hectare along with process bioethanol yields. Forest residues are 20 percent of the overall harvested timber and the collection percentage is 50 percent, meaning that 10 percent of total standing biomass is delivered to the thermochemical conversion facility as convertible biomass (Allen et al. 2005). Forest residues yield ranged from 11 to 25 OD tonnes/ha depending on the management intensity over a rotation period of 25 years. Additionally, annual bioethanol yield per acre varied from 43 to 94 liters/ha. At the production scale considered in this article, somewhere between 22,660 and 50,350 hectares would need to be harvested (and residues collected) per year for a biomass supply of 700,350 OD tonnes/y. This is similar to productivity values from previous studies (Evans and Cohen 2009, Somerville et al. 2010, Gonzalez et al. 2012).

Life-cycle stages.—Using SimaPro, data from the USLCI database, and the Aspen Plus simulation, GHG emission factors and fossil fuel usage factors were calculated for each scenario and for each chemical, biomass, energy, and waste stream. These factors were then multiplied by the quantity of each stream to determine the GHG emissions or fossil fuel–based energy used. Energy usage factors are listed in Table 4 for bioethanol and in Table 5 for gasoline. Emission

Table 3.—Biomass production rates and land use per unit of bioethanol produced based on yields as provided in the ''Biomass productivity" section.^a

| Management intensity | Total yield (OD tonnes/ha) | Annual yield (OD tonnes/ha/y) | Forest residues (OD tonnes/ha) | Forest residues collected (OD tonnes/ha) | Annual forest residue collection $(OD \tonnes/ha/v)$ | Ethanol land yield (liters/ha) | Annual ethanol land yield (liters/ha/y) |
|-------------------------|-------------------------------|----------------------------------|-----------------------------------|--|--|--------------------------------------|---|
| Low | 139 | 5.6 | 27.8 | 13.9 | 0.56 | 5.832 | 233 |
| Medium | 229 | 9.1 | 45.7 | 22.9 | 0.91 | 9.594 | 384 |
| High | 309 | 12.4 | 61.9 | 30.9 | .24 | 12.980 | 519 |

 $^{\text{a}}$ OD = oven dry.

Table 4.—Bioethanol cradle-to-grave fossil energy usage.

| Process | MJ per | Unit | Amount | MJ energy |
|---|--------------|---|--------|--|
| Raw materials Feedstock | 217 | tonnes | | $9.20E - 05$ $1.99E - 02$ |
| transportation Biomass gasification 7.80E-04 Fuel transportation Total | 1.19 1.19 | tonnes \times km 1.46E-02 MJ fuel tonnes \times km 2.73E-03 | 1.00 | $1.73E - 02$ $7.80E - 04$ $3.24E - 03$ $4.13E - 02$ |

factors are listed in Table 6 for bioethanol and Table 7 for gasoline.

Impact assessment

Global warming potential.—Using output from SimaPro and Aspen Plus, the GHG emissions for both gasoline and bioethanol were calculated. The GHG emissions are listed in kilograms of $CO₂$ equivalents per megajoule of fuel produced and consumed in a standard ignition passenger vehicle (Fig. 3).

When comparing the conventional gasoline and bioethanol fuel life-cycle GHG emissions, two product stages have the greatest impact on net life-cycle GHG emissions: the conversion process and residue growth. The biomass-tobioethanol conversion process life-cycle stage contributed the largest quantity of GHGs $(0.115 \text{ kg } CO₂)$ eq per MJ bioethanol produced) to the overall bioethanol life-cycle burdens. These emissions were due to the energy-intensive conversion processes that require large inputs of heat and power. In the analyzed scenario, the energy requirements are met through the combustion of raw syngas for combined process heat and power production. In the growth of the biomass, 0.169 kg CO₂ eq per MJ of fuel are captured and stored as plant matter; thus the emission factor for biomass growth was 0.169 kg $CO₂$ per MJ. This negative emission, or credit, greatly reduces the overall emissions to a net lifecycle emission of $2.28E-2$ kg $CO₂$ eq per MJ of bioethanol. This is a 74 percent reduction in $CO₂$ equivalents as compared with the conventional gasoline life cycle. Raw materials, transportation of raw materials, bioethanol fuel transportation, and fuel combustion emissions were all similar for both gasoline and bioethanol (von Blottnitz and Curran 2007, Ravindranath et al. 2009, Roberts et al. 2010).

These findings are consistent with previous studies surrounding the environmental burdens of biofuels production from forest biomass (Bright and Strømman 2009, Hsu et al. 2010, Daystar 2011).

Acidification.—Acidification is defined by the TRACI impact assessment method as the ''potential to cause wet or dry acid deposition'' (Bare et al. 2003). Acidification results from nitrogen dioxide (NO_x) and sulfur dioxide (SO_x) combining with water vapor in the atmosphere to form nitric

Table 5.—Gasoline cradle-to-grave fossil energy usage.

| Process | MJ per | Unit | Amount | MJ energy |
|--|---------------------------|------------------------------|------------------------------|--|
| Raw materials Feedstock | 1.02 | MJ gas | 1.00 | 1.02 |
| transportation Biomass gasification $7.22E - 02$ Fuel transportation | $2.44E-02$ MJ gas 1.19 | MJ gas tonnes \times km | 1.00 1.00 $1.68E - 03$ | $2.44E - 02$ $7.22E - 02$ $2.00E - 03$ |
| Total | | | | 1.12 |

Table 6.—Bioethanol cradle-to-grave emissions factors and global warming potential.

| Process | $kg CO2$ per | Unit | Amount | kg CO ₂ /MJ |
|--------------------------------|--------------|--------------------|---------------|---------------------------|
| Raw materials | 15.81 | tonnes | $9.20E - 0.5$ | $1.45E - 03$ |
| Uptake during growth | $-1,833$ | tonnes | $9.20E - 0.5$ | $-1.69E - 01$ |
| Feedstock | | | | |
| transportation | $9.32E - 02$ | tonnes \times km | $1.46E - 02$ | $1.36E - 03$ |
| Biomass gasification | 0.115 | MJ fuel | 1.00 | 0.115 |
| Fuel transportation | $932E - 02$ | tonnes \times km | $2.73E - 03$ | $2.54E - 04$ |
| Fuel combustion | $7.38E - 02$ | MJ $(LHV)^a$ | 1.00 | $7.38E - 02$ |
| Total | | | | $2.28E - 02$ |

 a LHV = lower heating value.

and sulfuric acid in dilute concentrations. These acids are then reintroduced to the troposphere and various fragile ecosystems in the form of acid rain following the regional weather patterns. This study analyzed the acidification potential of both bioethanol and the energy equivalent quantity of conventional liquid gasoline fuel on a cradle-tograve basis (Fig. 4). In the analyzed biomass conversion process scenario, NO_x and SO_x are emitted during transportation, conversion chemicals manufacturing, on-site biomass combustion as smokestack emissions, and as tailpipe emissions during blended fuel combustion in light-duty passenger transportation (Bright and Strømman 2009, Cherubini and Ulgiati 2010).

The production and use of 1 MJ of bioethanol resulted in a 72 percent decrease in acidification emissions as compared with the gasoline scenario. The gasoline fuel production and raw material transportation resulted in significantly higher acidification emissions than the same bioethanol life-cycle stages. The emissions used to calculate the impacts of biomass conversion to bioethanol (fuel production) were generated using a process simulation because large-scale commercial production facilities do not currently exist from which operational data can be used. As a result of this uncertainty, emission flow rates will need validation as production processes come online.

Eutrophication.—Eutrophication is the primary impairment of surface water quality due to nutrient loading. Nitrogen and other nutrients enter the environment through either point sources (a localized quantifiable source) or nonpoint sources (undefinable sources difficult to quantify) and increase cyanobacteria growth. Once the cyanobacteria die, they settle to the bottom of the waterway and decompose, consuming dissolved oxygen (DO) and deplet-

Table 7.—Gasoline cradle-to-grave emissions factors and global warming potential.

| Process | $kg CO2$ per | Unit | Amount | kg CO ₂ /MJ |
|-----------------|--------------|--------------------|--------------|------------------------|
| Raw materials | 0.199 | kg | $2.29E - 02$ | $4.56E - 03$ |
| Raw material | | | | |
| transportation | $6.78E - 02$ | kg | $2.86E - 02$ | $1.94E - 03$ |
| Fuel production | $5.28E - 03$ | MJ | 1.00 | $5.28E - 03$ |
| Fuel transport | $9.32E - 02$ | tonnes \times km | $1.68E - 03$ | $1.57E - 04$ |
| Fuel combustion | $7.45E - 02$ | MJ | 1.00 | $7.55E - 02$ |
| Total | | | | $8.74E - 02$ |
| | | | | |

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Figure 3.—Cradle-to-grave global warming potential comparison of bioethanol and gasoline. Also shown is the scenario in which some of the burdens of forest management are allocated to the bioethanol (AL). Total values are shown above the bar graphs.

Figure 4.—Cradle-to-grave acidification impacts of bioethanol and gasoline. Also shown is the scenario in which some of the burdens of forest management are allocated to the bioethanol (AL). Left axis displays individual life-cycle stage acidification impacts and the right axis displays the total acidification impacts. Total values are shown above the bar graphs.

Figure 5.—Cradle-to-grave eutrophication impacts of bioethanol and gasoline. Also shown is the scenario in which some of the burdens of forest management are allocated to the bioethanol (AL). Total values are shown above the bar graphs.

ing the total DO level in the waterway. In extreme cases this results in fish kills (Sharpley et al. 2003).

This study used the TRACI impact assessment method to quantify eutrophication in terms of N equivalents for a cradle-to-grave scope (Fig. 5). Eutrophication for the bioethanol case was 59 percent lower than for gasoline. Eutrophication due to gasoline production and use was largely a result of atmospheric emissions of nitrous oxide and other nitrogen-containing airborne emissions. Because the bioethanol production process uses less fossil-based fuel, the raw material, raw material transportation, and fuel production stages had lower eutrophication impacts. Airborne emissions from the modeled thermochemical conversion process are based on simulation results because no current industry data exist. In our base-case scenario, fertilizer use emissions were allocated 100 percent to the timber and pulpwood product life cycles. Thus, no eutrophication from fertilizer use was attributed to the forest residues–to–bioethanol conversion scenario. This assumption is explored in a sensitivity analysis in a later section.

Cherubini and Ulgiati (2010) suggested that biofuels often have a higher eutrophication potential than a conventional fossil fuel life cycle. Unlike our study, Cherubini and Ulgiati assumed full burdens for the bioethanol scenario from biomass production, which resulted in higher eutrophication and acidification impacts than with conventional fossil fuels.

Fossil fuel input

In some biofuels scenarios (e.g., corn-derived bioethanol) the fossil fuel requirements to produce the biofuel result in small net fossil fuel savings compared with gasoline or other fossil fuels (Kikuchi et al. 2009). In this study, fossil fuel usage was tracked from cradle-to-grave for the bioethanol and gasoline scenarios. Fossil fuel energy usage was reported as megajoules of fossil fuel consumed per megajoule of transportation fuel produced (Fig. 6).

The fossil fuel energy usage during gasoline production was determined to be 1.12 MJ/MJ of fuel produced according to the GREET model calculations. This is due in large part to the use of crude oil as a feedstock for gasoline production. This value is similar to previous studies analyzing fossil fuel input for gasoline production (Davis et al. 2009). Bioethanol production and use in this study was determined to require 0.041 MJ of fossil fuel energy per MJ of bioethanol. The majority of this fossil fuel usage resulted from biomass transportation and forest residue collection activities. Overall, a fossil fuel usage reduction of 96 percent was estimated for the production and use of bioethanol as compared with the gasoline alternative.

GREET results comparison

GREET 2011 was used to simulate an equivalent bioethanol fuel life cycle and calculate upstream and downstream environmental burdens. The results from the GREET model analysis were compared with the results of this report (Fig. 7), and both fuel production and feedstock production results were noticeably different. Through careful examination of the GREET and SimaPro model methodologies, burden allocation methods were found to cause much of the difference in results. Within the feedstock production stage, GREET allocates only a portion of the

Figure 6.—Nonrenewable energy inputs for production of 1 MJ of bioethanol (no burdens allocated) and gasoline fuels. Total values are shown above the bar graphs.

Figure 7.—Global warming potential of bioethanol from forest residues (no burdens allocated) as compared with standard Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation model values. Net values (triangles) are reported.

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carbon dioxide flux to the fuel and the rest to energy created within the biomass conversion process. Within this study, all carbon in the biomass supply was represented as a carbon credit and all emissions from the conversion process were attributed only to ethanol production. Additionally, the fuel production process within the GREET model is allocated negative GHG emissions (carbon credits) for electrical and process heat energy cogeneration from knots, fines, and waste biomass during the conversion process, thus reducing the net GHG emissions from this stage. The GREET gasification data were noted as ''uncertain,'' likely contributing to variation in the fuel GHG emissions. A sensitivity of the GREET versus SimaPro data validity is explored later in this study. Overall, GHG emissions were found to be slightly higher than the equivalent bioethanol thermochemical conversion scenario in GREET.

Allocation sensitivity

Forest residues are often considered as a waste or byproduct of forest operations, although it is acknowledged that residues can contribute to changes in soil and habitat quality through soil nutrient and carbon recharge after residue decomposition. The decay of residues above ground (emitting greenhouse gases) can sometimes be essentially complete. Thus, there is not yet a firm conclusion on how to allocate the burdens of the alternative end-of-life scenario (natural in-woods decomposition) for the forest residues. In many studies and LCA models, forest residues are not allocated any of the environmental burdens associated with the production and management of the main forest products (Wang 2001) based on the assumption that residues are waste material. It is of interest to compare the ''no-burden'' scenario as used in the body of this study with a scenario that allocates forest management burdens to the primary wood product and residues by mass fraction, ''allocated burdens.''

For this scenario, it is estimated that 20 percent of the total aboveground tree mass is considered to be forest residues and 50 percent of residues are collected (Allen et al. 2005). Thus, 10 percent of the aboveground tree mass is collected as residues and 80 percent is collected for the primary forest product. Then the mass percentage of collected residue to primary product plus collected residue is 11 percent, and this percentage is used to allocate the same percentage of feedstock development burdens to the collected residue and thus to the bioethanol from gasification conversion scenario. Reforestation emissions including land preparation, seedling production, planting, fertilization, pesticide application, and herbicide use are accounted for using USLCI data within the SimaPro LCA software. TRACI impact assessment methods were used to determine global warming potential, acidification, and eutrophication from cradle-to-grave. The gasoline burdens were calculated using SimaPro for well to pump and using the GREET model for combustion emissions.

The difference in global warming impacts for the allocated-burdens bioethanol scenario was minimal (Fig. 3). The acidification burden increased between the noburden and the allocated-burden scenarios by approximately 27 percent (Fig. 4). Eutrophication in the allocated-burden scenario increased by about 210 percent compared with the no-burden scenario (Fig. 5), primarily because of forest fertilization. Fertilizers have the potential to run off into streams as well as to volatilize into nitrogen compounds, ultimately resulting in nutrient loading as a nonpoint source. Interestingly, the eutrophication in the allocated-burden scenario was about 26 percent higher than gasoline, in stark contrast to the no-burden result, which showed 146 percent lower eutrophication for bioethanol compared with gasoline.

Study limitations

The goal of this study was to analyze the GHG emissions and energy requirements for the production and use of bioethanol produced from forest residue indirect gasification to mixed alcohols. In performing this analysis, necessary assumptions were made to equalize the systems being analyzed and to isolate production variables for better comparison. One significant assumption was that only pinebased forest residues are input into the conversion facility as biomass feedstock. In all likelihood, more than one feedstock type would be used to maximize biomass availability and to reduce life-cycle costs by lowering transportation distances, increasing feedstock flexibility, and reducing operational risk due to potential supply chain logistics issues (Gonzalez et al. 2012). The thermochemical conversion process has been shown to be flexible enough to accept various biomass sources, which changes with changing feedstock prices, harvesting schedules, locations, and other logistical constraints. Despite this limitation, isolation of pine forest residues as the only feedstock was necessary for this study so life-cycle impacts could be calculated in an interpretable and consistent manner, which is useful when quantifying the effect of biomass feedstock growth and collection on the biofuels production process net burdens.

Additionally, the assumption that combustion of ethanol produced from forest residues would result in biogenic (i.e., carbon neutral) atmospheric emissions impacts the outcome of the study as a result of a large reduction in net GHGs associated with the bioethanol production life cycle using this accounting methodology.

Finally, if forest residues were assumed to decompose entirely to solid organic matter increasing soil carbon, the removal of this forest residue would impact the long-term carbon stock in the soil and would increase net GHG emissions.

Conclusions

For the forest residues–to–bioethanol thermochemical conversion pathway modeled in this study, biomass growth and fuel production life-cycle stages had the largest GHG emissions. Biomass growth contributed a relatively large negative GHG emission to the overall GHG emissions value, offsetting to some extent the positive GHG emissions released in the fuel production and fuel combustion stages. Both raw material and fuel transportation were found to have a minimal impact on net life-cycle GHG emissions. Bioethanol produced from thermochemical conversion of forest residues qualifies as an advanced biofuel under the Renewable Fuels Standard, having a net GHG reduction of 74 percent compared with gasoline. Fossil fuel usage per megajoule of transportation fuel was 96 percent lower for bioethanol than for gasoline on a cradle-to-grave basis. The fossil energy usage of gasoline was higher mainly because of the nonrenewable feedstock input (crude oil). Acidification and eutrophication were significantly lower for the bioethanol scenario than the gasoline scenario when forest operation burdens were not allocated to the forest residues. With a mass-based allocation of forest management burdens to the forest residues scenario, eutrophication was higher

than for the gasoline fuel scenario. This was mainly due to the fertilizer emissions value allocated to residue growth and harvest. Global warming potential and acidification were not as sensitive to the method of allocation of forest management burdens as eutrophication.

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