U.S. DEPARTMENT OF ENERGY

Summary of Fast Pyrolysis and Upgrading GHG Analyses

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December 2012

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Acknowledgments

Acronyms and Abbreviations

ANL	Argonne National Laboratory
EISA	Energy Independence and Security Act
GHG	greenhouse gas
HHV	higher heating value
INL	Idaho National Laboratory
LCA	life cycle analysis
LHV	lower heating value
MYPP	multi-year program plan
NETL	National Energy Technology Laboratory
NREL	National Renewable Energy Laboratory
PNNL	Pacific Northwest National Laboratory
RFS2	Renewable Fuel Standard 2
SOT	state-of-technology

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1.0   Introduction

The Energy Independence and Security Act (EISA) of 2007 established new renewable fuel categories and eligibility requirements (EPA 2010). A significant aspect of the National Renewable Fuel Standard 2 (RFS2) program is the requirement that the life cycle greenhouse gas (GHG) emissions of a qualifying renewable fuel be less than the life cycle GHG emissions of the 2005 baseline average gasoline or diesel fuel that it replaces. Four levels of reduction are required for the four renewable fuel standards. Table 1 lists these life cycle performance improvement thresholds.

Table 1. Life Cycle GHG Thresholds Specified in EISA

Fuel Type	Percent Reduction from 2005 Baseline
Renewable fuel	20%
Advanced biofuel	50%
Biomass-based diesel	50%
Cellulosic biofuel	60%

Notably, there is a specialized subset of advanced biofuels that are the cellulosic biofuels. The cellulosic biofuels are incentivized by the Cellulosic Biofuel Producer Tax Credit (26 USC 40) to stimulate market adoption of these fuels. EISA defines a cellulosic biofuel as follows (42 USC 7545(o)(1)(E)):

> The term "cellulosic biofuel" means renewable fuel derived from any cellulose, hemicellulose, or lignin that is derived from renewable biomass and that has lifecycle greenhouse gas emissions, as determined by the Administrator, that are at least 60 percent less than the baseline lifecycle greenhouse gas emissions.

As indicated, the Environmental Protection Agency (EPA) has sole responsibility for conducting the life cycle analysis (LCA) and making the final determination of whether a given fuel qualifies under these biofuel definitions. However, there appears to be a need within the LCA community to discuss and eventually reach consensus on discerning a 50–59 % GHG reduction from a ≥ 60% GHG reduction for policy, market, and technology development. The level of specificity and agreement will require additional development of capabilities and time for the sustainability and analysis community, as illustrated by the rich dialogue and convergence around the energy content and GHG reduction of cellulosic ethanol (an example of these discussions can be found in Wang 2011).

GHG analyses of fast pyrolysis technology routes are being developed and will require significant work to reach the levels of development and maturity of cellulosic ethanol models. This summary provides some of the first fast pyrolysis analyses and clarifies some of the reasons for differing results in an effort to begin the convergence on assumptions, discussion of quality of models, and harmonization.

2.0   Review and Analysis

Recently, several organizations have undertaken LCAs of GHGs for biofuels from the fast pyrolysis of biomass and upgrading of bio-oil. The results of these LCAs are summarized in Table 2. Directly

comparable results for forest residue are highlighted in red in row 1 (Pyrolysis Gasoline GHG Emissions) and row 5 (Reduction from Baseline) of Table 2. These results range from 62–68% reduction in GHG emissions from the petroleum baseline.

Table 2. Summary of GHG Results for Fast Pyrolysis and Upgrading - Woody Biomass

	1	2	3	4	5	6	7	8	9	10	11
	NREL/PNNL Forest Res. Grid Power, NG[1] Apr 2011 MYPP	NREL/PNNL Forest Res. Biomass Power, NG[1] Apr 2011 MYPP	NREL/PNNL Forest Res. Grid Power, NG[1] Nov 2011 MYPP	NREL/PNNL Poplar Grid Power, NG[1] Nov 2011 MYPP	ANL/PNNL Forest Res. Grid Power, NG[1] Nov 2011 MYPP	UOP/Ensyn Logging Res. Grid Power, No NG (py-oil for H_2)	UOP/Ensyn Poplar Grid Power, No NG (py-oil for H_2)	UOP/Ensyn Willow Grid Power, No NG (py-oil for H_2)	GHGenius Wood Res.[2,3] Grid Power, NG[1]	GHGenius Short Rotation Forestry Crop[2,3] Grid Power, NG[1]	GHGenius Standing Timber[2,3] Grid Power, NG[1]
Pyrolysis Gasoline GHG Emissions, gCO_2-e/MJ	38.9 (49 before revisions - published in Hsu 2011)	25.0 (35 before revisions - published in Hsu 2011)	31.5	36.8	35.8	29[4]	23	21			
Pyrolysis Diesel GHG Emissions, gCO_2-e/MJ[5]									30.1	42.4	37.2
Petroleum Gasoline GHGs, gCO_2-e/MJ[6]	93.4	93.4	93.4	93.4	93.4	91.2	91.2	91.2			
Petroleum Diesel GHGs, gCO_2-e/MJ[7]									95.4	95.4	95.4
% Reduction from Baseline	58%[8]	73%[8]	66%	61%	62%[9]	68%	75%	77%	68%	56%	61%
Notes on Assumptions	2 co-products: gasoline & diesel; process chemicals included; 50% feedstock moisture	2 co-products: gasoline & diesel; process chemicals included; 50% feedstock moisture	2 co-products: gasoline & diesel; 30% feedstock moisture	2 co-products: gasoline & diesel; 30% feedstock moisture	2 co-products: gasoline & diesel; 30% feedstock moisture	3 co-products: gasoline, diesel, & kerosene	3 co-products: gasoline, diesel, & kerosene	3 co-products: gasoline, diesel, & kerosene	Single product, transportation fuel blending stock	Single product, transportation fuel blending stock; includes land use changes (direct) and fertilizer	Single product, transportation fuel blending stock
Reference	Hsu 2012; Hsu 2011	Hsu 2012; Hsu 2011	This report	This report	Han et al. 2011	Kalnes et al. 2009	Kalnes et al. 2009	Kalnes et al. 2009	(S&T)2 Consultants 2011	(S&T)2 Consultants 2011	(S&T)2 Consultants 2011

[1] NG is natural gas. Conversion stage based on PNNL design report and associated data (Jones et al. 2009).

[2] GHGenius results are originally presented in higher heating value (HHV) and converted to lower heating value (LHV) (using HHV/LHV=1.07).

[3] GHGenius presents results for *combined* gasoline and diesel fuels, and compares them to conventional petroleum diesel fuel.

[4] Higher because of longer transportation distances assumed for logging residue. No feedstock chemicals or growing inputs, however.

[5] Diesel results are generally representative of the gasoline results and therefore are not presented for all of the studies.

[6] The GREET 2005 conventional gasoline baseline is 93.4 gCO_2-e/MJ (486 gCO_2-e/mile). The National Energy Technology Laboratory (NETL) gasoline baseline is 96.3 g/MJ-LHV (Skone and Gerdes 2009). The UOP/Ensyn and GHGenius studies have also evaluated petroleum baseline GHGs and are listed in the corresponding columns.

[7] The GREET 2005 conventional diesel baseline is 96.3 g CO_2-e/MJ (416 g CO_2-e/mile). The NETL baseline for diesel is 95.0 g/MJ-LHV (Skone and Gerdes 2009). The UOP/Ensyn and GHGenius studies have also evaluated petroleum baseline GHGs and are listed in the corresponding columns.

[8] The GREET 2005 conventional gasoline baseline is 93.4 gCO_2-e/MJ and 302 g CO_2-e/km (486 gCO_2-e/mile). NREL compares their final result per distance travelled on pyrolysis gasoline, 117 gCO_2-e/km (188 gCO_2-e/mile), to the reference value and reports a 61% reduction. However, NREL assumes a fuel economy of 25.4 mpg, instead of what GREET uses as the 2005 value, 23.4 mpg. Therefore, when comparing NREL's g/MJ value to the GREET baseline, NREL obtains 58%, but when comparing on the basis of g/mile, NREL obtains 61%. This is an inconsistency to be aware of and that merits resolution at a later date. One way to make a change would be to change the fuel economy to 2005 standards, or recalculate the GREET baseline for g/mile, using the 25.4 mpg.

[9] Current default case included in GREET 2011 for forest residue pyrolysis gasoline uses an external H_2 source, and therefore has higher GHG emissions and lower emission reductions relative to the case presented here (55%).

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Initially, the National Renewable Energy Laboratory (NREL) published a study in April 2011 (Hsu 2011) using conversion data provided by Pacific Northwest National Laboratory (PNNL) from the 2017 design case (Jones et al. 2009). Shortly thereafter, NREL and PNNL worked together to refine the LCA model, resulting in several revisions to the model and GHG results. These results (both before and after the revisions) are given in Table 2, column 1, and NREL recently published them in a journal article (Hsu 2012). NREL also evaluated a biomass-based electricity case, shown in Table 2, column 2. Cases given in columns 1 and 2 assumed feedstock moisture and preprocessing power from the U.S. Department of Energy's April 2011 Biomass Multi-Year Program Plan (MYPP) (DOE 2011a), which are listed in Table 3.

Meanwhile, Argonne National Laboratory (ANL) conducted a study (Han et al. 2011) also using conversion data provided by PNNL for the 2017 design case. In addition, PNNL provided the new assumptions used for the November 2011 Biomass MYPP (DOE 2011b) technical tables for feedstock moisture content and preprocessing power developed by Idaho National Laboratory (INL), as shown in Table 3. The results from ANL's study are given in Table 2, column 5. Table 3 compares the new feedstock assumptions (included in the November 2011 MYPP) with the previous assumptions from the April 2011 MYPP. PNNL helped thoroughly review and refine the conversion data used for ANL's GREET model[1] (ANL 2011) at a one-day meeting at ANL and in follow-up correspondence prior to the rollout of the updated version of GREET.

Table 3. Forest Residue Feedstock Moisture and Preprocessing Assumptions

	Old Front-End (Apr. 2011 MYPP)	New Front-End (Nov. 2011 MYPP)
Feedstock Moisture	50%	30%
Feedstock Moisture after Drying	7%	10%
Grinding and Drying (Fan) Power, MW	8.1	4.8
Process Offgas used for H2, lb/hr	4,902	10,863

PNNL then updated the revised NREL SimaPro model for the forest residue 2017 design case with INL's new preprocessing assumptions (see Table 2, column 3). Figure 1 shows the gasoline GHG emissions, natural gas usage, and electricity usage (per megajoule of gasoline and diesel fuel produced) for the 2017 design case using the April 2011 and November 2011 MYPP feedstock assumptions. The new feedstock moisture and drying assumptions result in lower drying energy and, ultimately, lower natural gas usage and associated GHG emissions for the system. This is because less system offgas (approximately 10 times less) is required to provide heat for feedstock drying (in addition to that from char combustion), and the offgas can instead be used to replace natural gas in the hydrogen plant. In addition, the new lower estimates for feedstock grinding and drying result in lower electricity requirements and hence lower related GHG emissions.

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[1] More information on the GREET model can be found at http://greet.es.anl.gov.
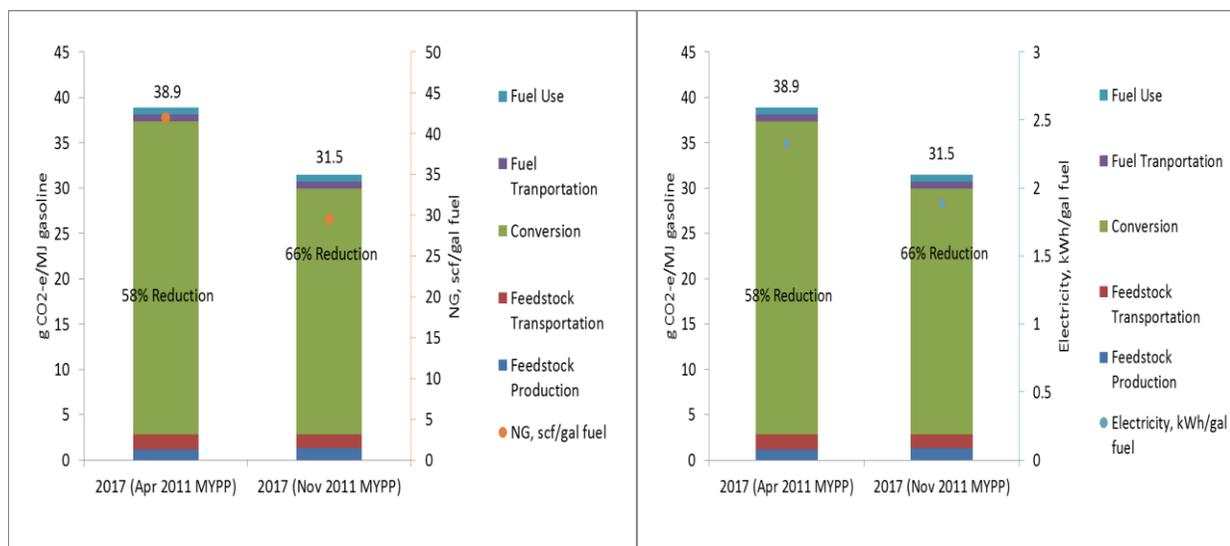
Note: Conversion stage includes feedstock preprocessing. Percent reduction is relative to GREET 2005 petroleum gasoline baseline, 93.4 gCO$_2$-e/MJ.

Figure 1. Pyrolysis gasoline GHGs, natural gas use, and power consumption (import) for 2017 target case (forest residue) using April 2011 MYPP and November 2011 MYPP assumptions for feedstock moisture, drying, and preprocessing power.

PNNL also recently modeled the 2017 design case with hybrid poplar, the feedstock assumed for the pathway in the November 2011 MYPP. The resulting GHG emissions from this analysis are given in Table 2, column 4. Emissions for hybrid poplar are higher than those for forest residue because the energy required for cultivation and harvesting of a purpose-grown crop does not apply to a waste product such as forest residue. Poplar cultivation and harvesting inventory for this model was developed by NREL, based on data for short rotation willow from the literature (Inman 2012; Abrahamson et al. 2002; Felix et al. 2008; Hartsough and Spinelli 2002; Spinelli and Hartsough 2006; Volk T. 2010). However, the energy consumption values for harvesting equipment (feller, skidder, delimber, and chopper) were revised using data from INL (Searcy and Hess 2010). Energy for transportation and handling were assumed to be the same as in the NREL/PNNL study of forest residue (Table 2, column 3).

Other studies of life cycle GHGs for fuels from fast pyrolysis and upgrading using woody biomass include those by UOP/Ensyn (Kalnes et al. 2009) and the Canadian LCA model GHGenius ((S&T)[2] Consultants 2011). UOP/Ensyn studied their rapid thermal processing (RTP) process using three different woody feedstocks (Kalnes et al. 2009), the results of which are given in Table 2, columns 6–8. It is important to note that for these cases, UOP/Ensyn assumes no natural gas consumption, but instead a cut of pyrolysis oil is used to provide all upgrading hydrogen, which may at least partially account for the lower GHGs relative to the other studies. The publication (a presentation) for this study provides limited details, and it is not clear as to what feedstock moisture is assumed. GHGenius also analyzes three woody feedstocks (GHGenius 2011; (S&T)[2] Consultants 2011), using data provided by Ensyn for the fast pyrolysis process and PNNL data (Jones et al. 2009) for the upgrading process. The GHGenius results (Table 2, columns 9–11) are provided for the composite gasoline/diesel fuel and are compared to conventional petroleum diesel in the report.

ANL also analyzed fast pyrolysis and upgrading fuels from corn stover feedstock using conversion data from a recent NREL report, the results of which are presented in Table 4 (Wright et al. 2010).

Similar to the UOP/Ensyn process, this study considers a case where a cut of the pyrolysis oil is used to provide hydrogen for hydrotreating and hydrocracking, thereby avoiding fossil GHG emissions associated with steam reforming of natural gas and greatly reducing the fuel's carbon footprint. It should be noted that process technology for reforming of bio-oils is still in the early stages of development.

Table 4. GHG Results for Fast Pyrolysis and Upgrading – Corn Stover

	ANL (GREET) – Corn Stover, Grid Electricity and NG[1]	ANL (GREET) – Corn Stover, Grid Electricity and Py-oil for H_2[1]
Pyrolysis Gasoline GHG Emissions, gCO_2-e/MJ	30.3	13.2
Petroleum Gasoline Baseline GHGs, gCO_2-e/MJ[2]	93.4	93.4
% Reduction from Baseline	68%	86%

[1] Conversion stage based on design by Iowa State, ConocoPhillips, and NREL (Wright et al. 2010).
[2] The GREET 2005 conventional gasoline baseline is 93.4 gCO_2-e/MJ (486 gCO_2-e/mile).

Results from analyses having similar assumptions for feedstock type and the conversion process are the most comparable and are shown in red in Table 2. These include the forest residue cases from the NREL/PNNL (column 3), ANL/PNNL (column 5), and GHGenius (column 9) models. These studies all assume a forest residue feedstock, the use of grid electricity, and the use of supplemental natural gas for hydrogen. (The NREL/PNNL study using April 2011 MYPP feedstock assumptions is excluded in lieu of the updated version.) GHG reductions from the petroleum baseline range from 62−68% for these studies. The differences between the estimates may be partially explained by breaking out the GHG contributions by stage of the fuel supply chain, as shown in Table 5. Note the ANL/PNNL values for individual stage contribution were estimated from graphical results (Han et al. 2011). The ANL/PNNL, NREL/PNNL, and GHGenius analyses for forest/logging residue (Table 5, columns 1–3) have similar fuel production stage GHGs; however, GHGs for feedstock production and transportation vary significantly. Estimates for the feedstock production stage range from 0–1.4 gCO_2-e/MJ and for feedstock transportation from 0.9–4.8 gCO_2-e/MJ. (NREL's value for fuel distribution, 0.7 g CO_2-e/MJ, was assumed for the ANL/PNNL analysis and was subtracted from the aggregate 5.7g CO_2-e/MJ for transportation.) Although these stages are relatively small contributors to the overall carbon footprint, differing assumptions for them can affect whether or not the fuel would qualify as "cellulosic" (60% or greater GHG reduction from petroleum baseline).

Table 5. GHG Stage Contribution for Fast Pyrolysis and Upgrading - Woody Biomass, CO_2-eq/MJ

	ANL/PNNL[1] Forest Residue, Grid Electricity and NG (Nov 2011 MYPP)	NREL/PNNL Forest Residue, Grid Electricity and NG (Nov 2011 MYPP)	GHGenius Wood Residues, (Canadian) Grid Electricity[2]
Feedstock Chemicals (fertilizer, herbicide)	0	0	0
Feedstock Production	1.4	1.3	0
Feedstock Transport	5.5[3]	1.6	0.9
Fuel Production	28.1	27.1	27.3
Fuel Transport		0.7	0.9[4]
Fuel Combustion	0.8	0.8	1.1
Total	35.8	31.5	30.1

[1] Estimated from graphical data (Han et al. 2011, Fig. 20).
[2] Results are presented for "Refined Bio-Oil," combined gasoline and diesel fractions.
[3] Includes feedstock and fuel transportation.
[4] Includes fuel dispensing, which is typically not included in other studies

Results in Table 2 shown in blue (columns 4 and 10) can also be appropriately compared. These results are for hybrid poplar feedstock (corresponding to the MYPP pathway) and a short rotation forestry crop, showing GHG reductions of 61% and 56% from the petroleum baseline, respectively. The remaining GHG estimates listed in Table 2 are not appropriately comparable to those previously discussed because they have significantly different assumptions regarding feedstock or feedstock conditions (as with cases in columns 1 and 10) or fuel sources for conversion (as with the NREL biomass power case in column 2 and the UOP/Ensyn cases that use pyrolysis oil for hydrogen generation).

Power and natural gas usage contribute the majority of GHGs to the fuel cycle (approximately 85% for the NREL/PNNL forest residue case, Table 2, column 3) and as such are critical to the analyses. The UOP/Ensyn study assumes a cut of bio-oil can be used to provide all hydrogen. As expected, this option results in lower conversion stage GHG emissions, but naturally at the cost of product yield. PNNL is involved in evaluations of both the economics and the GHGs of a modified version of the 2017 design case that uses pyrolysis oil instead of natural gas for hydrogen generation. Of particular interest is the identification of a "sweet spot," where both economics are favorable *and* GHG reductions are maximized.

To further illustrate the trade-off between GHGs and yield for this fuel pathway, Figure 2 shows results for the 2017 design case (Table 2, column 4), along with the 2011–2012 State-of-Technology (SOT)/Projection and 2013–2014 Projection cases. These cases are for hybrid poplar feedstock and correspond to the design and economics presented in the November 2011 MYPP. Advances in the technology over time are assumed to increase pyrolysis oil and product fuel yields from the current SOT to the target case. As fuel yields increase, lower amounts of offgas and char are available for hydrogen generation and heat and power generation, and higher yields of bio-oils require additional amounts of hydrogen for upgrading, both factors thereby increasing natural gas consumption and associated GHG emissions. Figure 3 shows the change in natural gas consumption. Plant power on an absolute basis is 13.0, 15.9, and 18.3 MW for the 2011–2012, 2013–2014, and 2017 cases, respectively. This increasing trend is the result of higher amounts of power for hydrogen compression (because more fuel is being

made) and lower amounts of heat from char available for steam and electricity generation. Note that power consumption per unit of fuel energy produced stays relatively constant over time because the same amount of hydrogen is used per unit of fuel produced (compression for which is the primary power consumer at the plant).

Use of forest residue is of particular interest for biofuel production because it is considered a waste product, and as such does not have the GHG emissions burdens of cultivating and harvesting a purpose-grown crop. Figure 4 shows life cycle GHGs using forest residue feedstock for the 2017 goal case (Table 2, column 3), along with the 2011–2012 and 2013–2014 projection cases. Note that this analysis assumes the same transportation distance (45 miles) for forest residue and hybrid poplar. This may not be the case in reality, as forest residue is expected to require a larger collection radius compared with farmed trees.
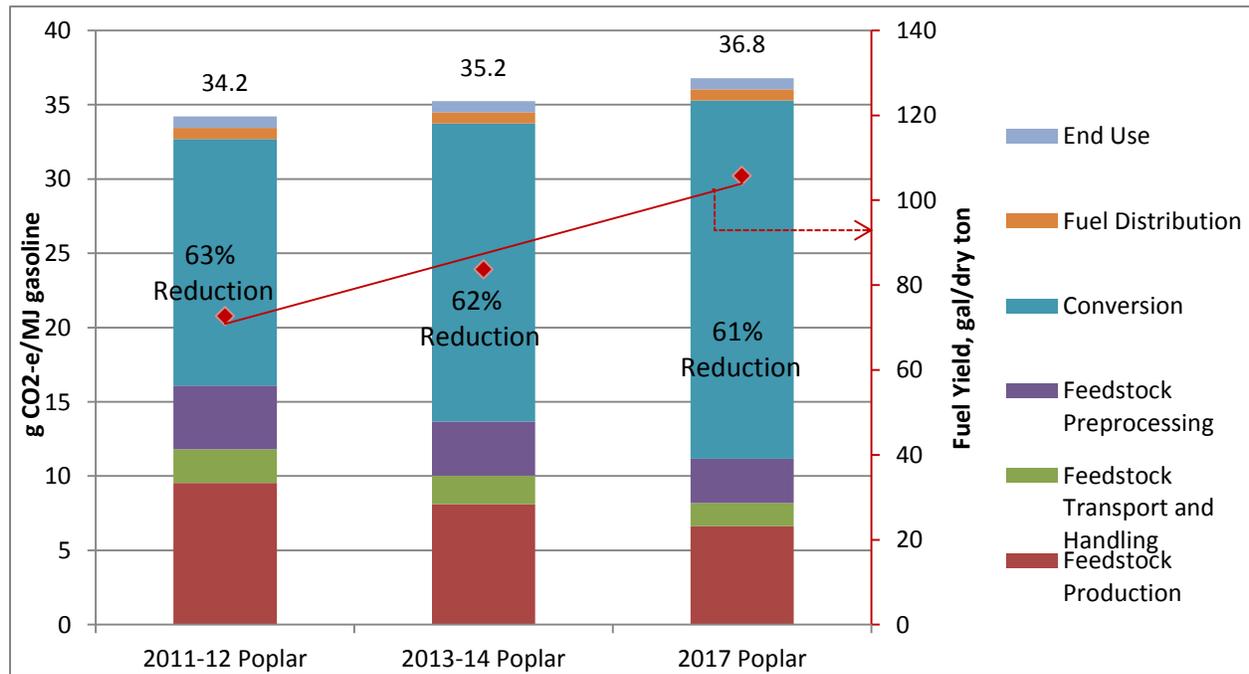
Note: Percent reduction is relative to GREET 2005 petroleum gasoline baseline, 93.4 gCO$_2$-e/MJ.

Figure 2. Pyrolysis gasoline GHGs, total fuel yield (gasoline and diesel), and GHG reductions using hybrid poplar (2011–2012 SOT/Projection, 2013–2014 Projection, and 2017 design case).
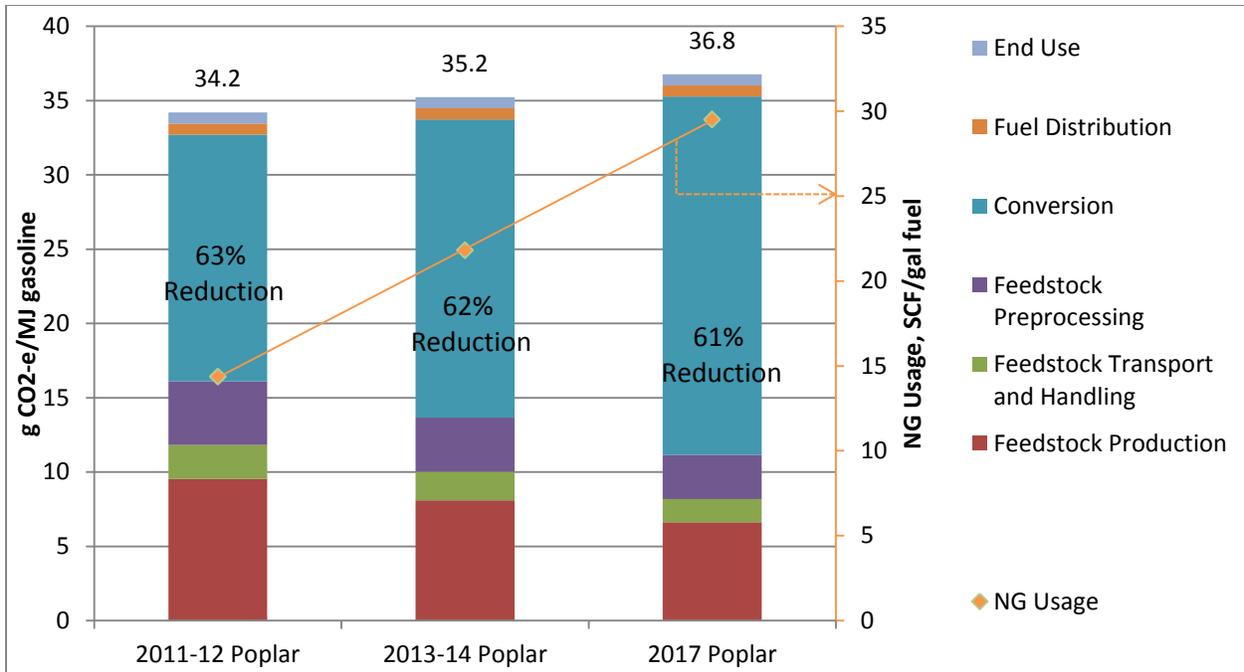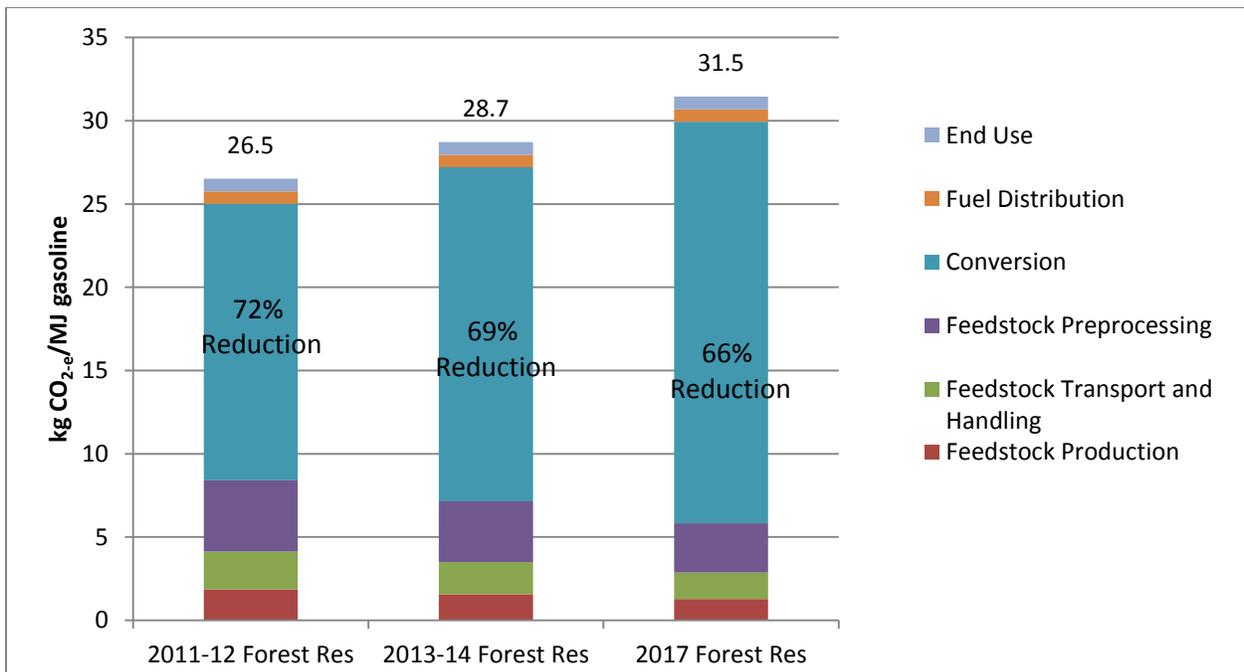
Note: Percent reduction is relative to GREET 2005 petroleum gasoline baseline, 93.4 gCO₂-e/MJ.

Figure 3. Pyrolysis gasoline GHGs and natural gas use for fast pyrolysis and upgrading using hybrid poplar.

Note: Percent reduction is relative to GREET 2005 petroleum gasoline baseline, 93.4 gCO₂-e/MJ.

Figure 4. Pyrolysis gasoline GHGs from fast pyrolysis and upgrading using forest residue.

3.0   Conclusions

From this initial review of recent GHG studies, the average value of estimates for forest residue (from the ANL/PNNL, NREL/PNNL, and GHGenius models) is 32.5 gCO_{2-e}/MJ, or a 65% reduction from the GREET 2005 petroleum gasoline baseline value (93.4 gCO2-e/MJ). Although these results indicate this fuel to be within the cellulosic biofuel definition range, only the EPA can make the final determination on whether this pathway qualifies under the EISA advanced or cellulosic definitions. Fewer estimates are available for hybrid poplar, though it is evident that GHG reductions will be less for a short rotation woody crop than a residue feedstock. However, further work on convergence in the larger community and clarification of the variance associated with the models would help clarify the GHG reduction classification boundary.

Technology advances that improve fuel yields may not always decrease GHG emissions, and in this specific instance may lead to higher GHG emissions, but may still be within the range of EISA's cellulosic biofuel definition. Reduced-fossil or non-fossil options, such as using biomass for power and hydrogen, will further reduce net GHGs for this pathway. However, the balance between economics (strongly influenced by yield) and GHGs needs to be fully understood and optimized. In addition, the technical feasibility of processes such as bio-oil reforming needs to be determined. Another important consideration is that the design case will be updated in the near future and will likely integrate new research on alternative approaches to direct liquefaction of biomass with processes including but not limited to catalytic pyrolysis, hydropyrolysis, and hydrothermal liquefaction. The revised design could therefore change significantly from its current state. Even with these changes, the balance between fuel product yield and GHGs will remain critical to the development and implementation of sustainable biofuels and is a key component of PNNL's analysis work for FY12 and beyond.

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         Name (#)

  #   Name
       Organization
       Address
       City, State and ZIP Code

  #   **Foreign Distribution**
      #   Name
         Organization
         Address
         Address line 2
         COUNTRY

  #   **Local Distribution**
       Pacific Northwest National Laboratory

Name	Mailstop
Name	Mailstop
Name	Mailstop
Name	Mailstop
Name	(PDF)