

Supporting Information

Techno-economic analysis and life-cycle analysis of two light-duty bio-blendstocks: isobutanol and aromatic rich hydrocarbons

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Aromatics and benzene blending limits

In order to reduce harmful emissions from vehicles, the EPA has introduced regulations that set maximum and/or minimum limits on gasoline fuel components. For example, EPA limits the amount of benzene allowed in gasoline to no more than 1.3% by volume of gasoline.¹ This requirement started under the MSAT (Mobile Source Air Toxics) gasoline program in January 2011. The MSAT provide guidelines to reduce hazardous air pollutants or air toxics emitted by cars and trucks.² California, on the other hand, implemented more stringent standards to those proposed by EPA under the MSAT program. For example, gasoline sold in California has even lower benzene levels and limits its content to 1.1% by volume.³ While EPA does not have a maximum limit on aromatics (i.e. benzene, toluene, ethylbenzene, xylenes, C9 and heavier and total aromatics), in California, the aromatics are limited to 35% by volume.³

Process flow diagram and process description of isobutanol

As shown in Figure S1, the process begins with raw lignocellulosic feedstock (20 wt% moisture) being fed to the plant. Feedstock handling at a satellite depot transforms the biomass into a uniform size (0.15-0.25 in) and then pelletizes the milled material. Once the biomass pellets reach the conversion facility, a conveyor transports the material to pretreatment and conditioning. In all, it costs \$84.45/ton (2014) to purchase and process the biomass to the throat of the pretreatment reactor.⁴ The plant processes 2,000 dry metric tons of biomass a day. Pretreatment steps follow the same methods as the NREL 2013 design report,⁵ but are described briefly here. Dilute sodium hydroxide (17 mg/g dry biomass) combines with the biomass in the pretreatment reactor at 80°C. Following the one hour deacetylation reaction, screens at the bottom of the reactor dewater the resulting mixture to 30 wt% solids. This removes solubilized acetate and

undesirable inert species. A vertical vessel steams the biomass to 160°C and impregnates it with sulfuric acid (9 mg/g dry biomass), and then, with a residence time of five minutes, the biomass deconstructs in a horizontal pretreatment reactor at a pressure of 5.5 atm. Ammonia conditioning raises the discharged pretreatment reactor product stream to a pH of 5.

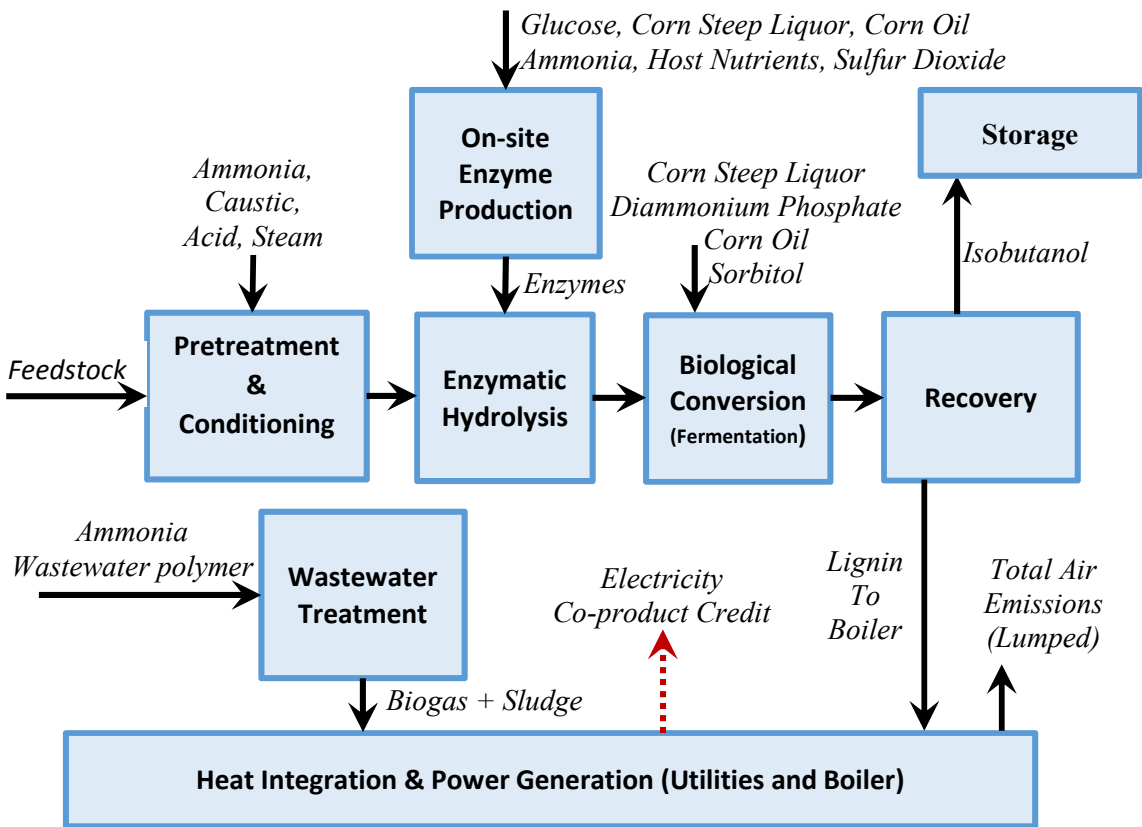


Figure S1 Isobutanol Process Flow Diagram

Similar to the pretreatment step, the enzyme production and enzymatic hydrolysis is based on the 2013 NREL design report and described in detail there.⁵ To summarize, the process creates cellulose enzyme on-site using seed trains with five 300 m³ aerobic fed-batch reactors. Target loading rates of protein to the enzymatic hydrolysis reactor are 10 mg protein/g cellulose. State-of-technology (SOT) loading rates have shown 12 mg/g cellulose for the enzymatic hydrolysis

reactor. Enzymatic hydrolysis converts cellulose to monomeric sugars. The hydrolysis starts with 20 wt% solids loading into eight continuous reactors (250,000 gallons). The continuous process takes 24 hours. Batch hydrolysis follows the continuous reactors and lasts 60 hours (3.5 days total). The SOT requires 5 days for the combined time of continuous hydrolysis and batch hydrolysis. In all, the conversion target case for hemicellulose to xylose is 90% and cellulose to glucose is 90%. Conversely, the SOT demonstrated conversion of hemicellulose to xylose is only 76% and cellulose to glucose of 74%. Dissimilar to more recent designs,⁵ the hydrolysis material does not go through solid liquid separation and sugar concentration before being fed to the fermentation reaction. Instead, similar to previous ethanol fermentation designs,⁶ we assume the fermentation can handle solids as well as lower sugar concentrations, and lasts 1.5 days.⁷ SOT Fermentation time is set at 65 hours.⁸

The slurry from the batch hydrolysis reactors enters anaerobic reactors (1,000,000 gal tanks) where it is inoculated with metabolically engineered *Escherichia coli* that can convert both C5 and C6 sugars to isobutanol liquor.⁷ The target case assumes that 95% of glucose, 85% of xylose, and 85% of arabinose go to isobutanol production. Cell-mass lactic acid and other minor co-products are also produced. SOT isobutanol fermentation from cellulosic materials operates at bench scale with low productivities (0.2 g/L/hr) with yields at 8% of glucose,⁹ and only recent work on xylose.¹⁰ However, GEVO argues that the productivities have reached commercial scale at 2 g/L/hr and 94% of the theoretical yield.⁸ GEVO has reported a current yield of 59 gallons of isobutanol per bone dry ton of woody biomass.⁸ We model the SOT case to have a yield of 59 gal isobutanol/ton biomass when pretreatment and enzymatic hydrolysis match target values. At lower pretreatment and enzymatic hydrolysis fermentable sugar yields, the isobutanol yield is 46 gal isobutanol/ton biomass.

Previous isobutanol process models,⁷ discussed the use of continuous vacuum stripping as a means to remove isobutanol while anaerobic conversion is occurring. Vacuum stripping has the benefit of maintaining isobutanol titers below 2 wt%, as at higher isobutanol concentrations the cells can be stunted or killed. Added benefits of vacuum stripping are that it does not require additional reactor volume to accommodate additional gases and can be done in tandem with the fermentation. The resulting streams from fermentation and vacuum distillation then undergo separation and purification. The vent stream from the anaerobic reactors, containing CO₂, enters a scrubber column that has additional water fed to it. The water recovers volatized isobutanol and returns it to the outlet liquid stream from the anaerobic reactors, which in turn is routed to a first distillation column. Isobutanol accumulates in the distillate in the first distillation column with water at a set ratio. Solids, remaining water, and other co-products coming out of the bottoms of the distillation column separate into solids and liquid streams via a solid-liquid separator. An on-site combustor uses the solids as feedstock, while the stillage water is routed to wastewater treatment for cleanup and ultimate reuse in the process.⁷ The boiler unit produces on-site heat and electricity for the process to uses. Any additional electricity beyond the demands of the process is sold to the grid at \$0.0572/kWh consistent with prior design reports.⁵ After the first distillation column, a decanter allows for the isobutanol and water to phase separate into an isobutanol rich stream and an aqueous rich stream. The aqueous stream returns to the first distillation column for higher isobutanol recovery rates. A second distillation column concentrates the isobutanol rich stream to 99 wt% with the bottoms being recycled back to the first distillation column. If co-products are produced above a specific threshold, then additional separation units may be required to reach a purified final isobutanol stream.

Process flow diagram and process description of aromatic-rich hydrocarbons (ARHC)

As shown in Figure S2, the process begins with woody biomass feedstock (10 wt% moisture) being fed to the plant. Feedstock handling at a satellite depot transforms the biomass into a uniform size and then pelletizes the milled material. Once the biomass pellets reach the conversion facility, a conveyer transports the material to the gasifier. In all, it costs \$84.45/ton (2014) to purchase and process the biomass to the throat of the gasification reactor. The plant processes 2,000 dry metric tons of biomass a day. Gasification through mixed alcohols production (primarily ethanol) is describe in detail in a previous study,¹¹ and is briefly described here.

Synthesis gas from biomass gasification in an indirectly heated gasifier is conditioned by tar reforming to convert tars, methane and light hydrocarbons to additional hydrogen and carbon monoxide. Wet scrubbing is then used to remove particulates prior to compression where the cooled syngas is pressurized to 3,000 psia. The pressurized syngas is converted primarily to ethanol and a very small amount of propanol and butanol in a down-flow type shell-and-tube reactor. The tubes are filled with a sulfide-type catalyst operating at 300° C. The reactor is operated isothermally; the heat of reaction is removed via steam generation on the shell side. This steam along with steam generated from gas cooling in the gasification and tar reforming sections are used to generate high pressure steam, which in turn is used for power generation and process heating.

The alcohol conversion process involves two catalytic steps. In the first step, the wet alcohol product is near completely converted at 350° C using a modified zeolite (V-ZSM-5 or InV-ZSM-5) catalyst followed by distillation to light gases, liquefied petroleum gas (LPG) and C₅₊. The C₅₊

127 fraction is composed of aromatics and highly branched hydrocarbons.¹² Higher alcohols are also
128 converted along with ethanol.¹³ A portion of the light gases and LPG is recycled around the first
129 reactor to convert the light olefins to additional C₅₊ product. C₄- olefins and paraffins not
130 converted in the first reactor are routed to a second reactor containing Ga-ZSM-5 catalyst
131 operating at 500° C to produce mixed aromatics.¹⁴

132 While the mixed aromatics from the second reactor could be further purified for chemical sales,
133 in this study they are assumed to be blended into the C₅₊ stream from the first reactor C₅₊ fuel
134 product. The alcohol-to-gasoline step and LPG conversion are in the experimental stages,¹²⁻¹⁴
135 and ethanol production has been demonstrated.¹¹ It should be noted that the ARHC process is
136 not limited to thermochemically derived alcohols, but is applicable to ethanol from any source
137 (e.g., biochemical production).

138 In the SOT case, a significant quantity of C₄ paraffins is formed, not all of which is converted to
139 liquid fuel in the second reactor. The second reactor produces a fair amount of benzene, which,
140 when combined with the C₅₊ fraction from the first reactor, limits the amount that can be blended
141 into the gasoline pool. The target case assumes that the first reactor catalyst can be tuned to
142 improve the selectivity towards more C₅₊, and that the second reactor catalyst can be formulated
143 to reduce the benzene content in the second reactor product.

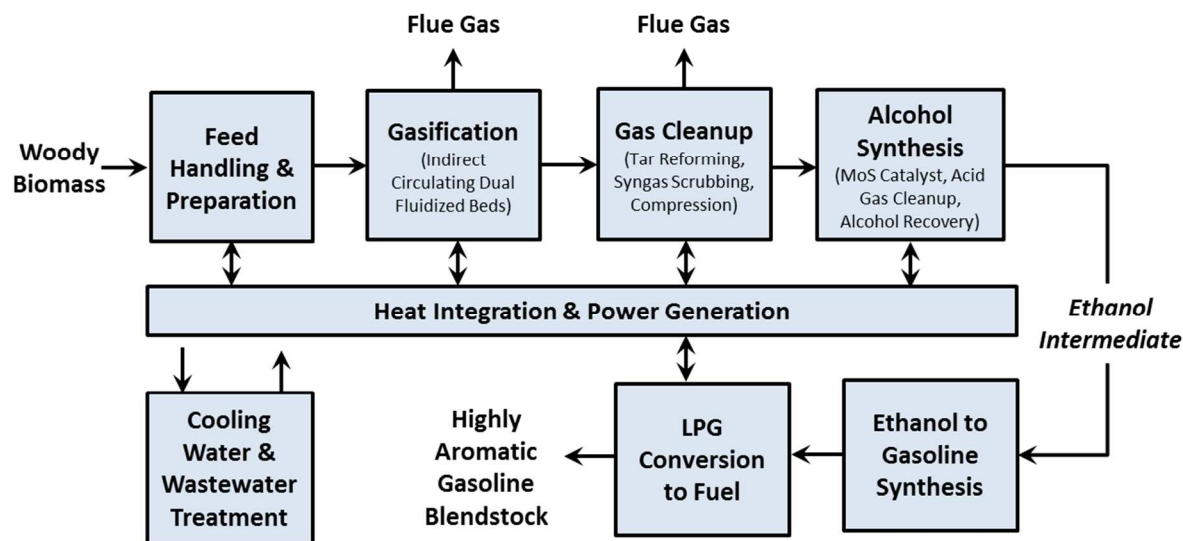
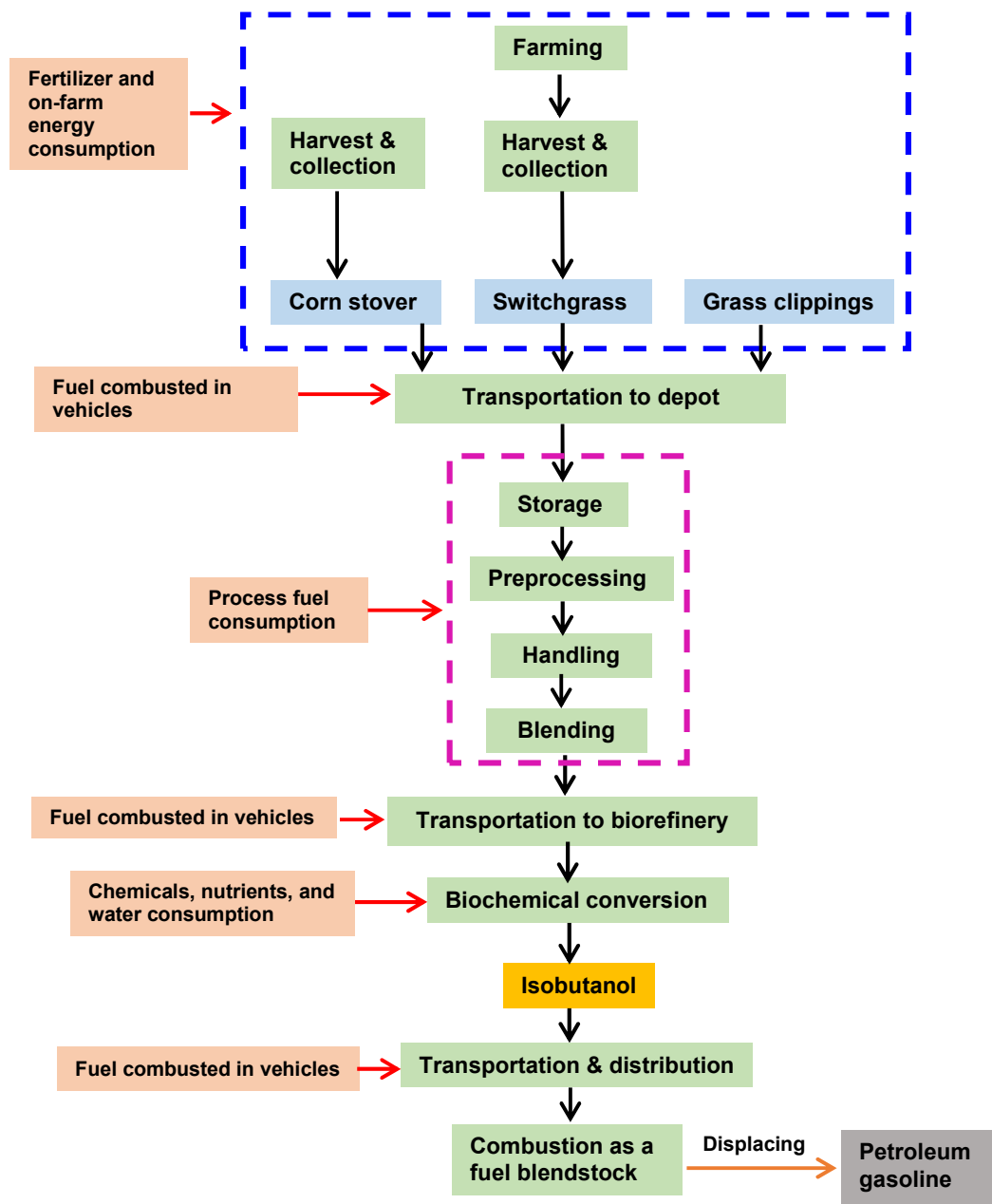


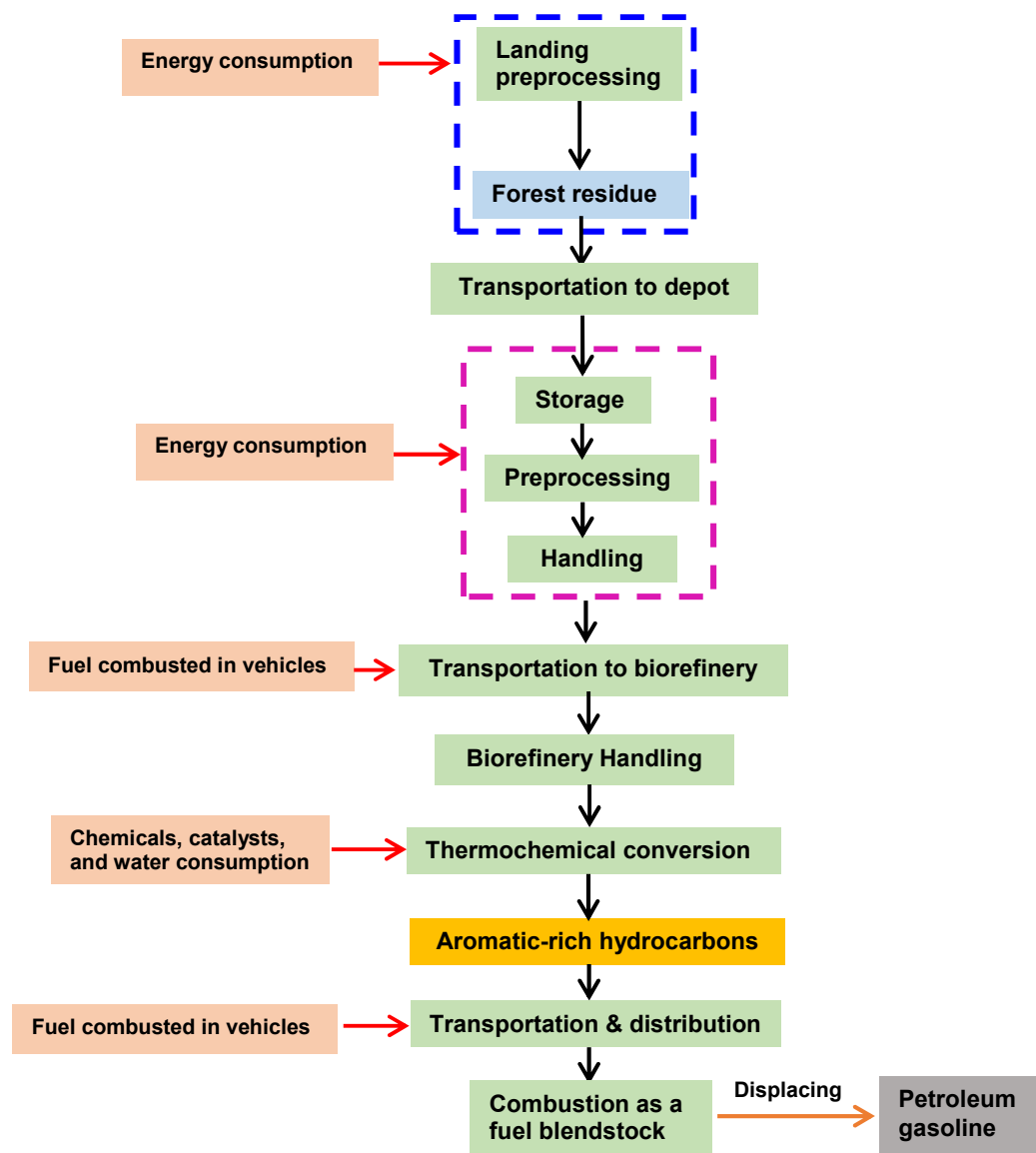
Figure S2 ARHC Process Flow Diagram

System boundary of life-cycle analysis

The LCA system boundary that considers the biomass feedstock supply chain, biorefinery operations, and transportation and end-use of isobutanol and ARHC is presented in Figure S3.

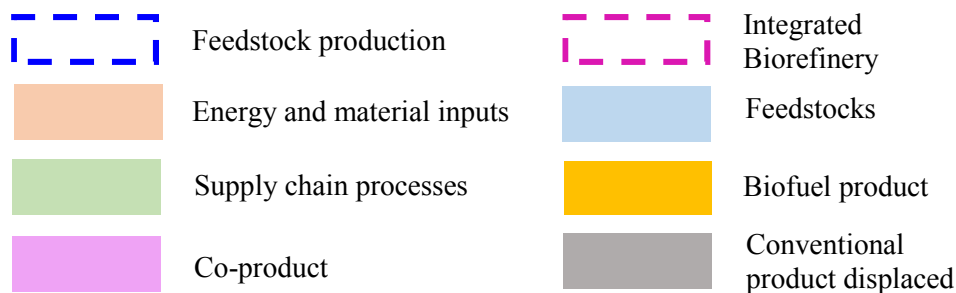


(a) Isobutanol



(b) Aromatic-rich hydrocarbons

Legend:



150

151 Figure S3. System boundary of life-cycle analysis of (a) isobutanol from biochemical conversion
 152 of herbaceous blend feedstock; and (b) aromatic-rich hydrocarbons from thermochemical
 153 conversion of forest residues.

Key data and assumptions about herbaceous feedstock blend and woody forest residues

Switchgrass fertilizer requirements and supplemental fertilizer requirements associated with corn stover were adopted based on GREET default values (see Table S1).^{15, 16} Data regarding energy consumption during switchgrass farming, harvesting, and logistic operation are also reported in detail in Table S2. A transition from three-pass corn stover harvest to two-pass harvest reduces feedstock logistics costs and contributes to both an improvement in the overall efficiency of the harvest operations and a reduction in ash content.¹⁷ The next logistics step is biomass storage. To enhance the biomass ease of conversion, the design case reduces storage-related dry matter losses of corn stover and switchgrass to 8% and 6%, respectively, through the minimization of microbial activity in storage, principally through controlled limitation of moisture content and/or oxygen in stored herbaceous feedstock. Processing of all feedstock components includes a preprocessing operation, which consumes mostly electricity. This operation includes size reduction, separation/sorting, drying, densification, ash reduction, and feedstock formation/blending. Details of fuel type and share for each feedstock preprocessing operation can be found in Table S3. Parameters used to determine energy consumed during feedstock transportation are shown in Table S4.

Table S1 presents switchgrass fertilizer requirements and supplemental fertilizer requirements associated with corn stover were adopted based on GREET default values.^{15, 16}

Table S1 Fertilizer usage, in gram/dry ton, of corn stover collection and switchgrass farming.^{15, 16}

Nitrogen (N)	Phosphate (P ₂ O ₅)	Potash (K ₂ O)	Calcium carbonate	Herbicides
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				(CaCO ₃)	
Corn stover	3,183	2,273	13,641	0	0
Switchgrass	4,877	2,308	3,200	5,847	53

Table S2 shows the energy consumption of production and logistics related to the herbaceous feedstock blend. Diesel consumption during switchgrass farming was taken from GREET default data,¹⁸ while harvesting and logistics operations are based on modeling by INL.¹⁷

Table S2 Energy consumption, in Btu/dry ton, for feedstock production and logistics

	Corn Stover (two-pass)	Corn Stover (three-pass)	Switchgrass	Grass Clipping
Farming ^{17, 18}			26,207	
Harvesting and Collection ¹⁷	108,560	89,040	41,270	
Storage ¹⁷	10,920	10,920	10,920	8,720
Preprocessing ¹⁷	236,870	236,870	318,000	317,190
Handling ¹⁷	8,230	8,230	7,750	19,500
Biorefinery handling ¹⁷	660	660	610	1,170

Table S3 presents the fuel type and share for each feedstock. As it is mentioned in the main paper, processing of all feedstock components includes a preprocessing operation, storage and handling. Electricity is the main source of energy for these unit operations.

Table S3 Share (%) of production and logistics stage fuel type for each feedstock.¹⁷

	Corn Stover (two-pass)	Corn Stover (three-pass)	Switchgrass	Grass Clipping
Harvest and Collection				

	Corn Stover (two-pass)	Corn Stover (three-pass)	Switchgrass	Grass Clipping
-Diesel	100%	100%	100%	-
Depot Preprocessing				
-Electricity	100%	100%	100%	100%
Depot Storage				
-Electricity	100%	100%	100%	100%
Depot Handling				
-Diesel	10%	10%	11%	14%
-Electricity	90%	90%	89%	86%
Biorefinery Handling				
-Electricity	100%	100%	100%	100%

188

189 Table S4 summarizes the key parameters of biomass transportation to a centralized depot and a
190 biorefinery.¹⁷ Logistics and fuel combustion emission data pertinent to the last two stages of the
191 supply chain, fuel transportation and distribution and fuel combustion, were obtained from
192 GREET.

193 Table S4 Feedstock transportation parameters for herbaceous blend.¹⁷

		Corn Stover (two-pass)	Corn Stover (three-pass)	Switchgrass	Grass Clipping
Transportation to depot	Truck payload (wet tons)	18	18	19	21
	Transportation distances (miles)	30	18	36	220
Transportation to biorefinery	Truck payload (wet tons)	21	21	21	
	Transportation distances (miles)	74	62	30	

194

195 There are several feedstock logistics operations for woody forest residues including landing
196 preprocessing/sorting, which consumes mostly diesel for steps of debarking, size reduction,

sorting, and screening. The transportation, depot storage, preprocessing, and handling stages consume mostly electricity. Energy requirements during feedstock logistics are shown in Table S5.

Table S5 Energy consumption, in Btu/dry ton, for forest residue logistics.¹⁹

	Forest Residues
Landing Preprocessing/Sorting	158,480
Depot Storage	8,720
Depot Preprocessing	482,950
Depot Handling	12,230
Biorefinery Handling	140

Table S6 presents the shares of fuel types related to the logistics of forest residues.

Table S6. Shares (%) of fuel types for forest residue logistics.¹⁹

	Diesel	Electricity
Landing	100%	-
Preprocessing/Sorting	100%	-
Transportation	100%	-
Depot Preprocessing		100%
Depot Storage	100%	-
Depot Handling		100%
Biorefinery Handling	-	100%

Parameters used to determine energy consumed during feedstock transportation are shown in Table S7. Vehicle payloads, transportation distance and moisture content, were informed by feedstock techno-economic analysis.¹⁹

Table S7. Feedstock transportation parameters

Transportation to depot	Truck payload (wet tons)	17
	Transportation distances (miles)	70
	Moisture content (wt%)	30%

Transportation to biorefinery	Truck payload (wet tons)	21
	Transportation distances (miles)	200
	Moisture content (wt%)	10%

Life-cycle analysis: Methodology to calculate NO_x and PM_{2.5} emissions

The production of bio-blendstocks and their end-use by vehicles involves combustion processes that produce NO_x and PM_{2.5} emissions. The process activities associated with bio-blendstock production pathways consume a diversified mix of process fuels by multiple combustion technologies with varying energy efficiencies and emission performances. For a given bio-blendstock, we applied a combustion technology-based approach to estimating the life-cycle NO_x and PM_{2.5} emissions of each life-cycle stage with the GREET model, using Equation S1.

$$LC_{CAP_{s,b}} = \left\{ \sum_p \sum_i \sum_j \left[\left(\frac{1}{\eta_p} - 1 \right) \times PF_{p,i} \times CT_{p,i,j} \times EF_{CAP_{s,i,j}} \right] + \sum_p \sum_i \left[Upstream_{CAP_{s,i}} \times \left(\frac{1}{\eta_p} - 1 \right) \times PF_{p,i} \right] \right\} + VO_{CAP_{s,b}} \quad (S1)$$

Where $LC_{CAP_{s,b}}$ is the life-cycle emissions of criteria air pollutant (CAP) s (either NO_x or PM_{2.5}), for bio-blendstock b (either isobutanol or ARHC); η_p is the energy efficiency of process p ; $PF_{p,i}$ is the share of process fuel i in process p ; $CT_{p,i,j}$ is the share of combustion technology j of process fuel i in process p ; $EF_{CAP_{s,i,j}}$ is the emission factor of CAP s for using process fuel i by combustion technology j (g/MJ); $Upstream_{CAP_{s,i}}$ is the upstream, or fuel-cycle emission of CAP s from production of process fuel i (g/MJ); and $VO_{CAP_{s,b}}$ are the vehicle tailpipe emissions of CAP s from vehicle operations (g/MJ).

Life-cycle analysis: Biorefinery-level results

Table S8 summarizes the biorefinery-level results of the target case, in comparison to the lower and higher fuel yield sensitivity cases, for ARHC. It shows that with higher fuel yield per ton of biomass converted, the total net emission reduction of the biorefinery increases, compared to the target and lower fuel yield cases.

Table S8. ARHC biorefinery-level GHG emission (in tons/year, assuming the biorefinery operates 350 days/year) results of the target case, in comparison to the lower and higher fuel yield sensitivity cases

	Target case	Lower yield	Higher yield
Biorefinery-level total emissions	158,515	122,551	184,917
Emission credit from displacing petroleum gasoline by ARHC	-579,948	-525,025	-636,733
Net emission reduction	-421,434	-402,474	-451,816

Key assumptions of sensitivity cases for TEA

Table S9 provides single-point sensitivity values that are considered in TEAs for isobutanol and ARHC. For Isobutanol, the assumptions include financial and cost values such as the return on investment, total capital costs, biomass cost, and interest rate. Other assumptions relate to the process model such as the conversion of xylose to isobutanol, enzyme loading, the cellulose to glucose for enzymatic hydrolysis, and fermentation time. For ARHC, financial assumptions, capital costs, operating costs and fuel yield were varied. For both bio-blendstocks, reasonable minima and maxima for each variable were chosen.

Table S9 Assumptions in the sensitivity analysis

Assumption Name	Min	Baseline	Max
Isobutanol			

Return on Investment	0%	10%	15%
Total CAPEX	80%	Baseline	140%
Conversion of Xylose to Isobutanol	90%	85%	50%
Enzyme Loading (mg/g)	5	10	20
Enzymatic Hydrolysis Cellulose to Glucose	95%	90%	75%
Biomass Cost (\$/ton)	\$75	\$84	\$90
Interest Rate	6%	8%	10%
FERM time (day)	1	1.5	3
ARHC			
Return on Investment	0%	10%	15%
Interest Rate	6%	8%	10%
Fuel Yield	110%	Baseline	90%
Biomass Cost (\$/ton)	\$75	\$84	\$90
Total CAPEX	80%	Baseline	140%
Syngas Compressor	80%	Baseline	140%
Gasifier and Reformer	75%	Baseline	125%
Mixed Alcohol Reactor	80%	Baseline	140%
Hydrocarbon Fuel	80%	Baseline	140%

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248 **Key assumptions of sensitivity cases for LCA**

249 Tables S10 and S11 provide process chemical, energy, and water consumption in sensitivity
250 cases for isobutanol and ARHC, respectively.

251 Table S10. Energy and material requirement, in gram, gallon, or mmBtu/mmBtu, for isobutanol production in the sensitivity cases, in
252 comparison to the target case

	Target Case	Low Enzyme Loading (5mg/g)	High Enzyme Loading (20mg/g)	Enzymatic Hydrolysis Cellulose Conversion to Glucose (75%)	Enzymatic Hydrolysis Cellulose Conversion to Glucose (95%)	Conversion of Xylose to Isobutanol (50%)	Conversion of Xylose to Isobutanol (90%)
Sulfuric Acid	4003	4003	4003	4426	3880	4605	3930
Caustic	2702	2702	2702	2987	2619	3108	2652
Ammonia	899	844	1011	1001	859	1180	866
Corn steep liquor	1828	1755	1999	1997	1775	2102	1794
Diammonium phosphate	213	215	215	236	207	245	209
Corn Oil	13	6	25	15	13	15	13
Glucose	2331	1167	4662	2577	2259	2681	2288
Polymer	6	6	6	6	6	7	6
FGD Lime	265	254	290	306	253	327	258
Makeup water consumption	70	68	74	66	86	92	68
Co-Produced Electricity	0.105	0.113	0.090	0.154	0.092	0.164	0.098

Table S11. Energy and material requirement, in gram, gallon, or mmBtu/mmBtu, for ARHC production in the sensitivity cases, in comparison to the target case

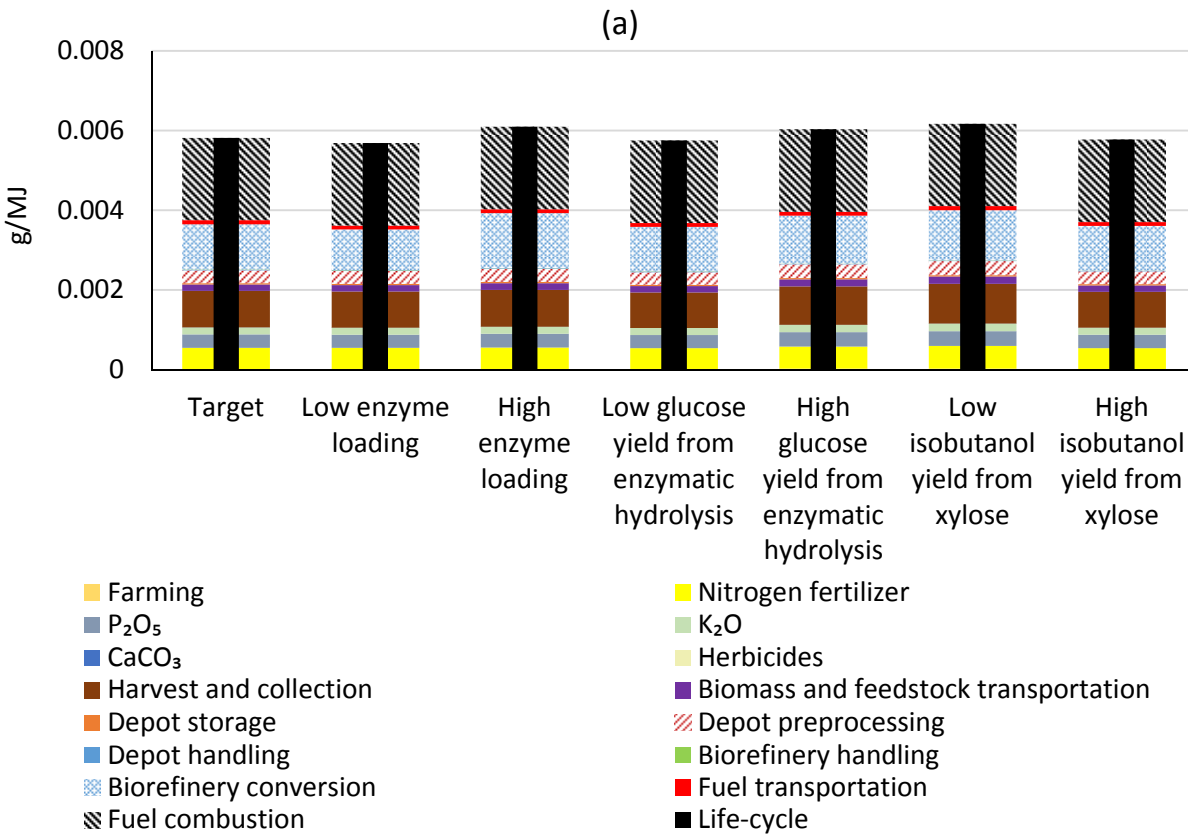
	Target	Lower Fuel Yield	Higher Fuel Yield
Fuel yield (mmBtu/dry ton of biomass)	7.5	6.8	8.3
Magnesium oxide	5.1	5.6	4.6
Fresh olivine	391.7	432.7	356.8
Tar reformer catalyst	8.7	9.7	8.0
Alcohol synthesis catalyst	29.1	31.7	29.5
Med Temp xx-ZSM-5 Catalyst	1.0	1.1	1.0
High temp xx-ZSM-5 Catalyst	0.7	0.6	0.6
DEPG solvent	1.3	1.4	1.2
Amine makeup	0.1	0.2	0.1
Diesel fuel	0.0	0.0	0.0
Makeup water consumption	50	54	30
External electricity	0.033	0	0.049
Diesel	0.002	0.002	0.002
Natural gas	0	0	0.074

PM_{2.5} emissions

For isobutanol in the target case, tailpipe PM_{2.5} emissions accounts for 36% of the total life-cycle emissions, followed by biorefinery conversion (20%), embedded emissions associated with production of fertilizers (18%), harvest and collection of herbaceous feedstocks by diesel-powered equipment (16%), and biomass depot preprocessing (6%). Life-cycle PM_{2.5} emissions in the sensitivity cases vary slightly because of relatively small variations in the emissions during the conversion step. As a result, all these cases have higher life-cycle PM_{2.5} emissions than those of petroleum gasoline.

For ARHC in the target case, emissions from fuel combustion accounts for 45% of the total, followed by landing preprocessing (25%), biomass depot preprocessing (11%), biomass transportation (8%), and biorefinery conversion (7%). The higher and lower fuel yield sensitivity

cases show a similar level of life-cycle $\text{PM}_{2.5}$ emissions as a result of tradeoffs between emissions from biomass conversion and biomass logistics that are impacted by the fuel yields. For both the isobutanol and ARHC pathways, tailpipe $\text{PM}_{2.5}$ emissions based on our assumption that these two bio-blendstocks have the same tailpipe NO_x emission factors as that of petroleum gasoline present the largest uncertainty due to lack of data about combustion emissions of isobutanol and ARHC in spark-ignition engines at this point. Further analysis is needed when information on emissions from engine combustion testing becomes available.



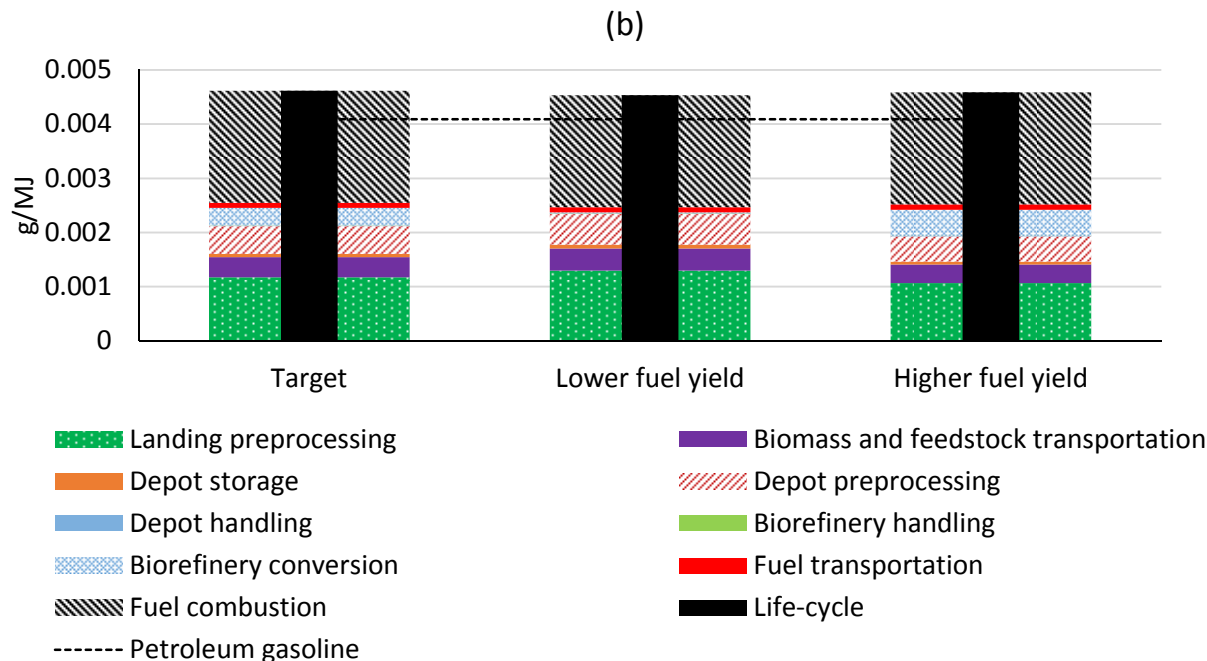


Figure S4. Life-cycle PM_{2.5} emissions of biomass-derived (a) isobutanol and (b) ARHC, in comparison to those of petroleum gasoline (solid black line)

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