

Supporting Information

Techno-economic analysis and life-cycle analysis of renewable diesel fuels produced with waste feedstocks

Longwen Ou^a, Shuyun Li^b, Ling Tao^c, Steven Phillips^b, Troy Hawkins^a, Avantika Singh^c, Lesley
Snowden-Swan^b, Hao Cai^{a,*}

^a Systems Assessment Center, Energy Systems Division, Argonne National Laboratory, 9700
South Cass Avenue, Lemont, IL 60439, United States

^b Pacific Northwest National Laboratory, 902 Battelle Blvd, Richland, WA 99354, United States

* Corresponding author, hcai@anl.gov, +1-630-2522892

^c National Renewable Energy Laboratory, 15013 Denver West Parkway, Golden, CO 80401,

United States

This Supporting Information has 33 pages, and contains:

Six Supporting Figures S1-S6

Twelve Supporting Tables S1-S12

Renewable Diesel from Swine Manure HTL

Process Description and Process Flow Diagram

As shown in Fig. S1, the process begins with the high moisture content swine manure collected from swine farms. Prior to processing, the swine manure is dewatered to 25 wt% solids for minimizing the capital and operating cost of the HTL plant. The swine manure cost at the gate of the HTL plant is assumed to be zero in the base case. However, there exist potential significant savings in avoided disposal cost to farms and transportation cost for the scenario collecting swine manure from multiple farms to support the HTL plant scale. The HTL plant processes 100 dry tonne of swine manure a day and is briefly described below.

The HTL reactor is operated at the subcritical water status with high solubility with organic compounds. First, the slurry swine manure is pumped to 20 MPa and heated in the heat exchanger and then trim heater to reach HTL reactor temperature of 350 °C. The HTL reactor has a shell-and-tube design with slurry in the tube side and hot heating oil in the shell side to maintain isothermal conditions. The organic matter in the feed is converted into biocrude, an aqueous phase with dissolved organic compounds, and a small amount of solids and gases. Solid is separated from liquid and gas using a filter and the filtered effluents are cooled for aqueous-biocrude-gas phase

separation. The HTL gaseous product combined with natural gas is sent to a burner for generating heat to supply the trim heater and HTL reactor via the hot oil system. The separated aqueous phase is sent to a series of treatment steps before recycling back to the WWT plant. First, it is treated with quicklime to raise the pH to ~11 and then stripped with air to remove ammonia and volatile organics (VOCs) in the aqueous stream. The removed ammonia and VOCs are destroyed in a thermal oxidizer (THROX) with the help of natural gas and catalyst. At the same time, the stripper bottom is pre-treated to decrease chemical oxygen demand (COD) before recycling back to WWT plants. Biocrude intermediate is cooled for storage and transported to the upgrader.

The HTL biocrude from multiple HTL plants is transported to a centralized upgrading plant. A transportation cost of \$0.02/GLE biocrude is assumed.¹ The main process steps of the upgrader include hydrotreating and hydrocracking. First, the biocrude feed is pumped to 10.5 MPa, mixed with compressed hydrogen, and preheated to the hydrotreater reactor temperature of 400 °C. The hydrotreating process can convert biocrude oxygen, nitrogen, and sulfur into CO₂ and water, ammonia, and hydrogen sulfide, respectively. The hydrotreater effluent is cooled to 25 °C to separate the gas and water and the resulting hydrocarbons are then fractionated into lights, naphtha, diesel, and heavy oil. Heavy oil combined with hydrogen is then hydrocracked at 400 °C and 7

MPa to produce additional naphtha and diesel blendstocks. Hydrogen is produced on site via steam reforming of the upgrading offgas and purchased natural gas.

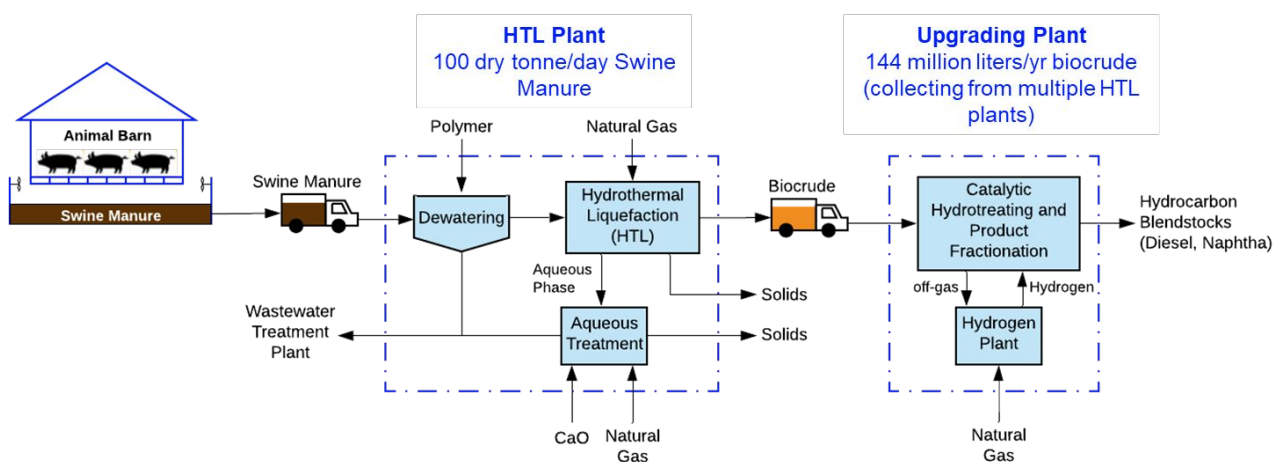


Fig. S1. Swine manure HTL and biocrude upgrading process flow diagram.

Swine Manure Feedstock Composition

Table S1 presents the ultimate analysis and proximate composition for swine manure tested in this work. The difference between experimental data and model input is mainly from data normalization and the ignored P.

Table S1. Ultimate and proximate composition of swine manure (75% moisture).

| Component | Experimental Data | Model Input |
|-----------|-------------------|---------------|
| | wt% dry basis | wt% dry basis |
| C | 47.6 | 47.1 |

| | | |
|---|------|------|
| H | 6.3 | 6.2 |
| O | 30.9 | 30.5 |
| N | 3.4 | 3.3 |
| S | 0.6 | 0.5 |
| Ash | 12.5 | 12.3 |
| P * | 1.4 | |
| * P is not modeled in the model due to the software limitation. | | |

Swine Manure Availability Distribution

Fig. S2 shows the swine manure distribution by production rate at the swine farms in the United States. The recoverable swine manure production rate per farm ranges widely, from < 0.5 dry tonne/day/farm to 225 dry tonne/day/farm.

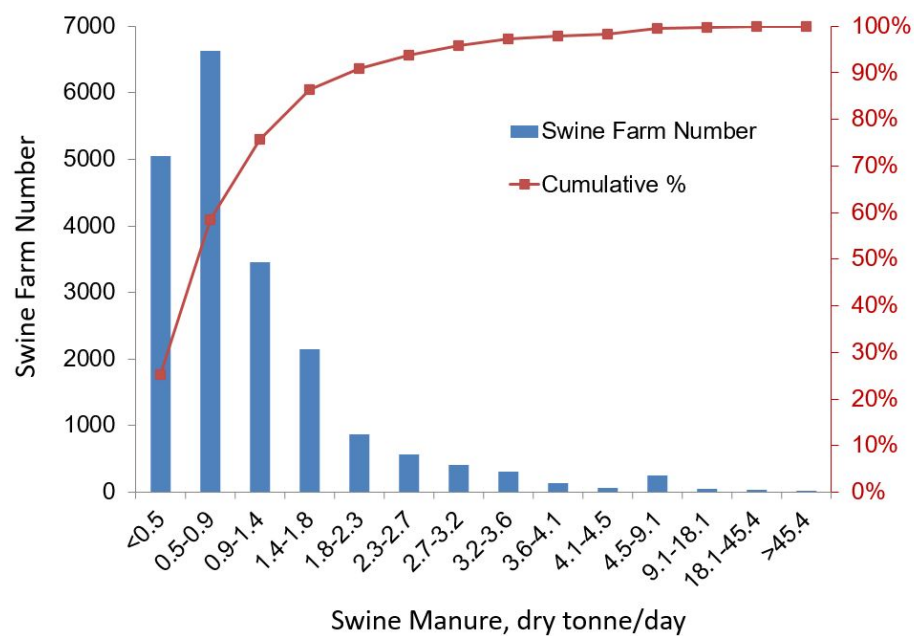


Fig. S2. Swine manure distribution by production rate at the swine farms in the United States.²

Swine Manure Transportation Cost

The transportation cost model developed by Marufuzzaman et al.³ is adopted to estimate the associated cost for transporting 25wt% solid swine manure 161 km per round trip (80-km collection radius). Table S2 lists the breakdown for transportation costs using facility-owned tandem trucks with a capacity of 40 m³. Fixed transportation cost is a function of trip number/year, which varies with total transported swine manure volume, truck utilization parameters such as loading/unloading time. A high yearly trip number can greatly reduce unit fixed cost (\$/km). Variable cost mainly includes the cost associated with fuel, labor, maintenance, and tire costs and thus unit variable cost stays the same for different HTL plant scales. The unit total cost is the sum of the fixed cost and variable cost. The trip cost can be calculated based on the assumed collection radius and the estimated unit transportation cost. As shown, the average unit transportation cost is about \$22 per dry tonne for the selected HTL plant scale from 27 - 227 dry tonne/day. Please refer to the literature³ for more detailed information on the transportation cost model and parameter values.

Table S2. Transportation cost estimation for different plant scales (25wt% solid, 161 km/round trip).

| HTL plant scale, dry tonne/day | Collection radius, km | Trip # per year | Fixed Cost | Variable cost | Total cost | Trip cost, \$ | Dry solid cost, tonne/trip | Unit cost \$/dry tonne swine manure | Average unit cost \$/dry tonne swine manure |
|--------------------------------------|-----------------------------|-----------------------|-------------------------|------------------|---------------|------------------|----------------------------------|--|--|
| | | | \$ /km wet swine manure | | | | | | |
| 27 | 80 | 948 | 0.38 | 1.19 | 1.57 | 253.1 | 10.48 | 24.16 | 21.59 |
| 227 | 80 | 7904 | 0.04 | 1.19 | 1.24 | 199.3 | 10.48 | 19.02 | |

Swine Manure Avoided Disposal Cost

In this analysis, the U.S. Department of Agriculture (USDA) method for estimating comprehensive nutrient management plans (CNMP) cost is used to approximate the avoided disposal cost for the swine manure treated by HTL plant, including on-farm nutrient management costs, off-farm transport costs, land treatment costs, manure and wastewater handling storage costs, and recordkeeping cost.⁴ The savings in commercial fertilizer costs due to CNMP implementation is considered as well by estimating the nutrient components value with an applied factor. Table S3 summarizes the estimated swine manure's CNMP implementation cost and nutrition value.

Table S3. Estimation of swine manure's CNMP implementation and nutrition value.⁴

| Farms # ¹ | Animal Units per farms ¹ | Total CNMP implementation Cost per farm ¹ , \$/yr | | |
|----------------------|-------------------------------------|--|------|-------|
| | | Average | Low | High |
| 32955 | 276 | 12029 | 2060 | 75159 |

| Total Solid kg/AU/day | Total Swine Manure dry tonne/yr | Swine manure nutrients ² | | |
|--------------------------------------|---------------------------------------|-------------------------------------|--------|------------------------------|
| | | N, wt% | P, wt% | applied factor |
| 3.98 | 13221585 | 5.4 | 1.44 | 0.4 |
| Total avoided disposal cost, mm\$/yr | | unit price ³ , \$/kg | | Nutrition credit, mm\$/yr |
| Low | High | N | P | |
| 67 | 2238 | 0.88 | 0.55 | 238 |

¹ based on the USDA report; ² PNNL test data; ³ USDA's historical database

Technology Readiness Level

MCCI Fuel from Yellow Grease to HEFA

HEFA from yellow grease has properties similar to conventional petroleum fuel, but the fuel has the advantages of a higher cetane number, lower aromatic content, lower sulfur content,⁵ and potentially lower GHG emissions. The HEFA conversion technologies are commercially available and are commonly used in today's refineries to produce transportation fuels. Neste Oil,⁶ Honeywell Universal Oil Products,⁷ Dynamic Fuels,⁸ and Diamond Green Diesel⁹ are a few leading companies producing HEFA fuel.

MCCI Fuel from Swine Manure HTL

HTL is a potentially viable, sustainable, and efficient pathway to convert wet wastes into drop-in diesel. The HTL biocrude can be upgraded via hydrotreatment to reach the conventional fuel properties with high diesel yield and high cetane number. The HTL technologies are still in

development and currently in the demonstration phase. The ongoing R&D from PNNL, Aalborg University, and pilot demonstration plants in Norway and Australia can bridge the knowledge gaps and help scale up the technology for commercialization.

Techno-economic Analysis Assumptions

MCCI Fuel from Yellow Grease to HEFA

Material and energy balance and flow rate information were generated using Aspen Plus¹⁰ process simulation software, assuming a feed rate to the biorefinery of 200,000 dry tonnes of yellow grease per year, which represents about 20% of the total annual yellow grease availability based on resource analysis performed by Milbrandt et al.² Data from process simulation were used to size and cost process equipment as well as compute raw material and other operating costs. The TEA model reasonably estimates a commercial-scale production cost of HEFA diesel. Table S4 summarizes key process model assumptions of the yellow grease to HEFA conversion.

Table S4. Key process model inputs for the yellow grease to HEFA pathway. Base case plant size processes 200,000 dry tonnes of yellow grease per year.

| Process variables | Model input |
|-----------------------|-------------|
| Feed solid (dry), wt% | 100 |

| | |
|---|-----|
| Feed impurity, wt% | 10 |
| Hydrocarbon fuel yield, GLE/dry tonne feedstock | 935 |
| Hydrotreating weight hour space velocity (WHSV), hr ⁻¹ | 0.5 |
| Hydrotreating catalyst life, year | 2 |
| Hydroisomerization and hydrocracking WHSV, hr ⁻¹ | 1 |
| Hydroisomerization and hydrocracking catalyst life, year | 5 |
| Catalyst costs, \$/kg | 134 |

MCCI Fuel from Swine Manure HTL

Table S5 lists the key process variables of the swine manure HTL process for this analysis, including the HTL feed solid and ash content, biocrude and hydrotreating fuel yield, the HTL reactor liquid hourly space velocity (LHSV), and hydrotreating catalyst weight hourly space velocity (WHSV). Increasing HTL feed solid content can improve the biocrude yield and reduce the capital and operating costs associated with processing feed water. However, the viscosity of the feed slurry increases with the higher solid content, making pumping potentially more challenging. A 25% solid feed was considered to be the upper bound of pumpable feed solid content based on the HTL bench-scale tests conducted at Pacific Northwest National Laboratory (PNNL).¹ The resulting biocrude yield was 50% (dry ash free (daf), wt%), which was higher than the target biocrude yield (48%) for sludge HTL.¹ This could be attributed to the high feed solid content, low ash content, and high fat content in the swine manure. The performance of

hydrotreater catalyst such as WHSV and lifetime has a great impact on the process economic projection. Currently, ongoing R&D is conducted to improve catalyst and reactor space velocity.

The values used in this model are consistent with the 2022 projected values¹¹ since this analysis is aiming to deploy the bioblendstocks after the target case technology is achieved.

It should be noted that HTL diesel usually has a relatively high cloud point (in the range of 4.3 – 5.1 °C) and must be blended with other blendstocks to produce on-spec diesel. Preliminary experimental data from PNNL suggests that a 30% blend level of the HTL diesel could achieve a cloud point of -15 °C, which surpasses the -10 °C required for No. 2 diesel. Therefore, it is expected that HTL diesel can be used in low blend level applications without the need for further processing to reduce its cloud point.

Table S5. Key process model inputs for swine manure HTL.

| Process variables | Model input | Experimental data ¹¹ |
|------------------------------------|------------------|---------------------------------|
| Feed solid (dry), wt% | 25 | 25 |
| Feed ash (dry), wt% | 12 | 12 |
| Biocrude yield (daf), wt% | 50 ^a | 49 ^a |
| Hydrotreated oil yield, wt% | 81 ^a | 84 |
| HTL reactor LHSV, hr ⁻¹ | 6 | 3.6 |
| Guard bed catalyst life, year | 0.5 ^b | 0.06 |

| | | |
|--|------|------|
| Guard bed WHSV, wt./hr per wt. catalyst | 1.3 | 0.42 |
| Hydrotreater catalyst life, year | 2 | 0.06 |
| Hydrotreater WHSV, wt./hr per wt. catalyst | 0.75 | 0.42 |

^a the model yield was adjusted slightly from experimental data to close the elemental balance.

^b lower than 2022 projected value due to high salts and metals in swine manure-derived biocrude.

Life Cycle Assessment Assumptions

GHG (CO₂, methane (CH₄), and N₂O) emissions, fossil energy consumption, water consumption, and emissions of two criteria air pollutants, NO_x and PM_{2.5} are the four environmental metrics assessed in this analysis. GHG emissions were calculated based on the 100-year Global Warming Potentials for CO₂, CH₄, and N₂O emissions, which are 1, 30, and 265, respectively.¹²

Methodology to Calculate NO_x and Particulate Matter (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 $\text{PM}_{2.5}$ emissions of each life-cycle stage with the GREET model, using Equation S1.

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

Where $LC_{CAP_{s,b}}$ is the life-cycle emissions of criteria air pollutant (CAP) s (either NO_x or $\text{PM}_{2.5}$), for bio-blendstock b (renewable diesel from either yellow grease or swine manure); 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).

Waste Management in the Swine Manure HTL Pathway

The swine manure HTL process produces aqueous and solid wastes that require treatment. For the aqueous phase waste, we consider a catalytic hydrothermal gasification (CHG) process to manage its chemical oxygen demand. The CHG process catalytically converts all organics to CO_2 and CH_4 .¹³ The CH_4 -rich off-gas from CHG is combusted to provide heat for the CHG reactor.

The carbon in the off-gas originates from the manure feedstock and thus the CO₂ emissions from combusting the off-gas to provide additional process energy required by the CHG process are considered biogenic, carbon-neutral, CO₂ emissions. Nitrogen available as dissolved ammonia in the treated wastewater can be stripped using quicklime and is considered in this analysis. In the case of sending the CHG treated wastewater containing ammonia to a wastewater treatment plant, we exclude any impacts in this analysis. The solid waste from the HTL process, which includes biochar, ashes, and residue biocrude, goes to landfill by truck for 80 km. The dry solid waste contains biochar and residual biocrude that accounts for about 26% of the dry solids. We assume that 13% of the dry solids are biochar. We assume that a carbon stability factor of 80%, an indicator of how much carbon in the biochar ends up sequestered in the soil after being applied to soil for 30 years,¹⁴ applies to the carbon in the solid waste upon landfilling. The remaining 20% of the carbon in the biochar and the carbon in the residual biocrude is assumed to be decomposed as CO₂ emissions, which are considered biogenic CO₂ emissions and do not contribute to the life-cycle GHG emission intensities of the MCCI bio-blendstocks. We account for the carbon sequestration effect of landfilling the solid waste. In addition, the solids from ammonia stripping are rich in CaCO₃, which presents an opportunity for the recovery of CaCO₃. In this analysis, we assume that

this solid stream goes to landfill. CaCO_3 in the solids could be acidified directly to CO_2 . We adopt a CO_2 emission factor of $0.216 \text{ g CO}_2/\text{g CaCO}_3$ for landfilling the CaCO_3 -rich solids from ammonia stripping based on an estimation by EPA.¹⁵

Counterfactual Scenario of Swine Manure HTL (Conventional Swine Manure Management)

Swine manure is high in nitrogen and moisture. In the counterfactual scenario, it requires proper management to prevent surface and groundwater contamination, protect the health of livestock and the public, and utilize manure nutrients for enhancing soil. Typical manure management practices involve storage, handling, treatment, and utilization to manage manure nutrients and achieve the above-mentioned goals. Major swine manure management systems currently adopted in the U.S. include deep pits, anaerobic lagoons, liquid/slurry storage, and applying to pasture as listed in Table S6. Recently, pig farms have been moving away from the lagoon and liquid storage systems to deep pits due to regulation.¹⁶

Table S6. Shares, CH_4 emission factors, and the fate of CH_4 -rich biogas of major swine manure management systems.¹⁷

| Swine Manure Management System | Deep Pit | Anaerobic Lagoon | Liquid/Slurry | Pasture |
|---|----------------|---------------------------|---------------------------|----------------|
| Management System Usage (MS%) | 69.9% | 15.5% | 13.4% | 1.3% |
| Manure Management System MCFs ($\text{CH}_4 / \text{CH}_4 \text{ max}$) ^a | 30.0% | 72.2% | 30.0% | 1.1% |
| Fate of CH_4 -rich biogas | 100% vented | 50% vented, 50% flared | 50% vented, 50% flared | 100% vented |

^a MCF: methane conversion factor. MCF is the ratio between the actual methane emissions and the maximum methane-producing capability for a swine manure management system. The maximum methane-producing capability for swine manure is 0.48 m³ CH₄/kg VS.¹⁷

Swine manure management practices could result in CH₄ emissions. These emissions vary by management systems, manure chemical composition, and climate.¹⁷ A fraction of the carbon in manure is converted to CH₄-rich biogas, which typically contains 60-70% CH₄.¹⁸ The CH₄ yields from these systems are listed in Table S6. The biogas may be emitted to the atmosphere, or flared, or collected as an energy source during manure treatment, while the rest of the carbon in manure ends up in the residue after treatment. Right now, only about 1% of the US pig farms install dedicated anaerobic digestion (AD) systems to collect and utilize CH₄-rich biogas for production of energy, such as electricity and heat.¹⁹ We assume that CH₄ emissions from anaerobic lagoons and liquid/slurry storage are manageable, but those from pasture and deep pit are not, due to technical challenges to install biogas collection systems.¹⁸ Given lack of information on how common the flaring practice is to mitigate fugitive CH₄-rich biogas, we assume that about 50% of the manageable biogas is flared while the rest become fugitive emissions to the atmosphere, and that all the technically unmanageable biogas become fugitive. For biogas flaring, we assume a flaring efficiency of 98%, i.e., 98% of CH₄ is combusted during flaring while the remaining 2% becomes fugitive emissions to the atmosphere. We note that presently most of the technically manageable biogas may not be flared and could become fugitive emissions because most pig farm owners would unlikely to install a biogas recovery system, which is an additional cost component, just to flare the recovered biogas. Therefore, our assumption that 50% of the biogas could be flared may be optimistic from a CH₄ destruction perspective. We evaluate the impact of a much less common flaring practice as part of the sensitivity analysis.

Solid residues after treatment contain nutrients such as nitrogen, phosphorus, and potassium, and can work as organic fertilizers for soil amendment, which replaces synthetic counterparts. We consider synthetic fertilizer displacement credits in the counterfactual scenario. N₂O is emitted when nitrogen is applied to soil by nitrification and denitrification, volatilization, and leaching.¹² However, N₂O emissions will also occur at the same rate when synthetic nitrogen fertilizer is applied.²⁰ Therefore, the net N₂O emission effect between the organic and synthetic fertilizers at

the application phase is zero. Nevertheless, organic fertilizers eliminate the need to produce synthetic fertilizers. Therefore, we account for energy and emissions from synthetic fertilizer production that would have been avoided in the counterfactual scenario as foregone benefits in the biofuel production scenario. The treated manure residues also contain a significant amount of carbon. The carbon in the residues is not stable and can be easily oxidized to CO₂. We assume that about 90% of the carbon in the solid residue is eventually converted to biogenic CO₂ after soil application, and the rest is sequestered and accounted for as CO₂ credits.²¹ We summarize energy consumption, nutrient contents, and carbon sequestration from land application of treated manure in Table S7.

Detailed carbon and nutrient flows in manure management are presented in Fig. S3. Table S1 shows the measured chemical composition of dry swine manure (i.e., total solids, or TS), which affects the carbon and nutrient balances before and after manure management. The emissions resulting from the counterfactual scenario can be avoided if manure is used for biofuel production that we evaluate in this study. We account for any avoided emissions from the counterfactual scenario as emission credits to biofuel production. Meanwhile, we account for foregone energy and emission benefits from applying treated manure as organic fertilizers to displace synthetic fertilizers manure that would have happened in the counterfactual scenario.

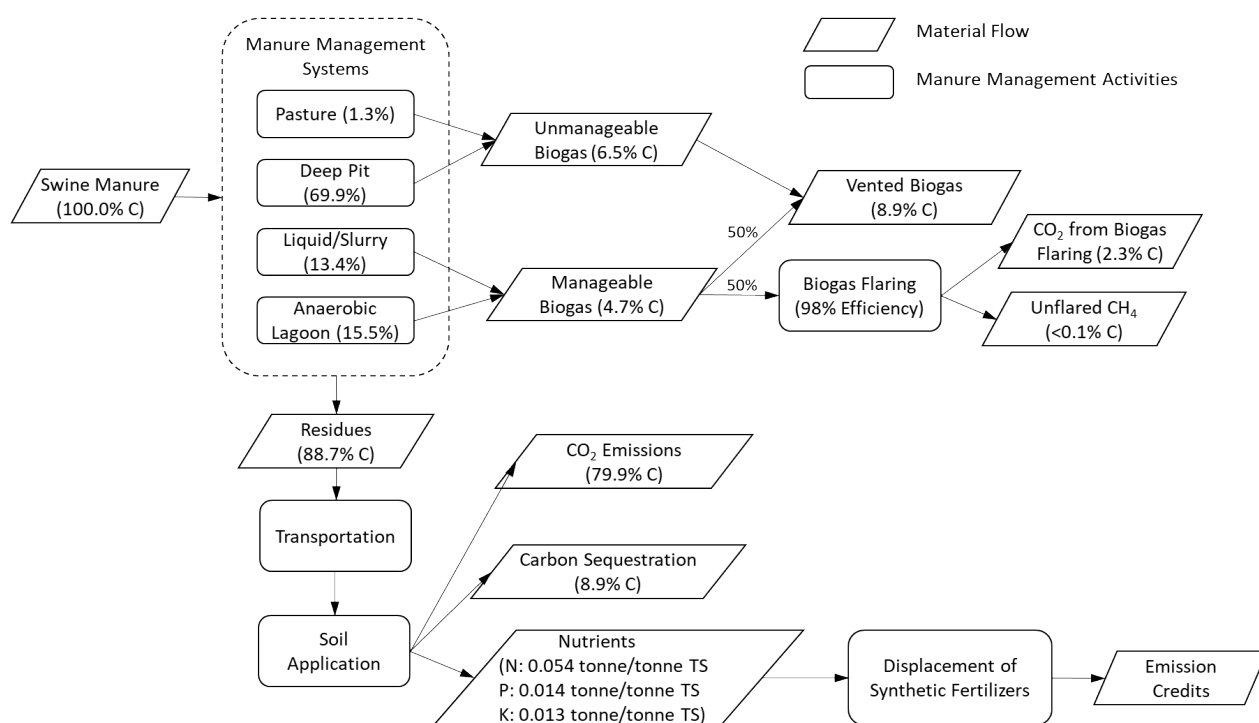


Fig. S3. System boundary and carbon balance of swine manure treatment in the counterfactual scenario. Notes: 1. The biogas generated from manure management systems consists of 65% of CH₄ and 35% of CO₂ by weight. 2. N₂O emission caused by soil application of treated manure is not included here because it is canceled out by the N₂O emission caused by the application of synthetic fertilizers that are displaced by the treated manure. 3. 98% of methane in the biogas is converted to CO₂, while the rest escapes to the atmosphere.

Table S7. Energy consumption, nutrient displacement credits, and carbon sequestration from land application of treated manure.

| | Parameter | Unit | Value |
|--|-----------|------|-------|
|--|-----------|------|-------|

| | | | |
|----------------------|--------------------|---------------------------|--------------------------|
| Energy consumption | Diesel | MJ/wet tonne | 3.0E+01 ²²⁻²⁴ |
| Nutrients | Nitrogen | kg/kg TS | 5.4E-02 |
| | Phosphorus | kg/kg TS | 1.4E-02 |
| | Potassium | kg/kg TS | 1.3E-02 |
| Carbon Sequestration | Sequestered Carbon | kg CO ₂ /kg TS | 1.4E-01 |

Table S8. MCF and energy consumption of a typical mixed plug-flow anaerobic digester.

| | |
|---|-------------------|
| Anaerobic digestion | |
| MCF of Anaerobic Digester (CH ₄ / CH ₄ max) ²⁵ | 81.7% |
| Electricity required for AD (kWh/wet tonne of manure) ²⁵ | 13.2 |
| Heat required for AD (MJ/wet tonne of manure) ²⁵ | 135 |
| Biogas cleanup | |
| NG processing efficiency ²⁶ | 94.4% |
| CH ₄ leakage ²⁵ | 1.0% ^a |

^a Han et al.²⁵ assumed 2.0% leakage of biogas during a two-stage cleanup and upgrading process

for renewable natural gas production. Since only one-stage cleanup is needed before biogas combustion for electricity generation, a leakage rate of 1.0% is assumed here.

Sensitivity Analysis Assumptions

MCCI fuel from yellow grease to HEFA

For economic assumptions, plant scale, feedstock costs, and capital costs are known to impact cost significantly. The optimistic plant scale of 300,000 dry tonne per year and the conservative scale of 25,000 dry tonne per year were both based on resource analysis performed by Milbrandt et al.² The ranges of feedstock price, total project investment (TPI), and hydrotreating capital cost were assumed $\pm 50\%$ of the base case value. In the base case, the MFSP was estimated without consideration of a fuel carbon credit under California's LCFS, which cultivates the largest renewable fuel market in the U.S. The sensitivity analysis investigates an optimistic case where the current fuel carbon credit of \$200/tonne CO₂ is considered,²⁷ assuming that the feedstock cost is influenced by the current fuel carbon credit under the LCFS. Hydrocarbon fuel yield often impacts the MFSP significantly. Since HEFA technology is relatively mature, a somewhat small variation in the fuel yield of $\pm 20\%$ was considered in an optimistic and a conservative scenario, respectively.

Table S9. Assumptions varied in the sensitivity analysis for yellow grease to HEFA pathway.

| Process Variables | Optimistic | Base Case | Conservative |
|-------------------|------------|-----------|--------------|
|-------------------|------------|-----------|--------------|

| | Scenario | | Scenario |
|---|----------|-----|----------|
| Economic assumptions: | | | |
| HEFA plant scale, thousand tonne/yr | 300 | 200 | 25 |
| Feedstock price, \$/dry tonne | 300 | 600 | 900 |
| Carbon credits, \$/tonne CO ₂ | 200 | 0 | - |
| Total project investment | -50% | | +50% |
| Hydrotreating capital cost | -50% | | +50% |
| Technical assumptions: | | | |
| MCCI hydrocarbon fuel yield (GLE/dry tonne) | 1122 | 935 | 747 |

MCCI fuel from swine manure HTL

Approximately 90% of the existing swine farms in the U.S. have a hog inventory of <5000 head/year and produce < 2.3 dry tonne/day manure. The largest industrial-scale swine manure farms (about 0.5% of swine farms in U.S.) produce from 9 to 227 dry tonne/day.² An HTL plant scale range of 27 to 227 dry tonne/day is selected considering the economies of scale as well as the recoverable swine manure production at farms. Although the majority of farms generate less than 27 dry tonne/day, swine farms in the United States are fairly centralized and about 70% of hogs are located in Iowa, Minnesota, North Carolina, Illinois, and Indiana and could be collected in areas within this region to potentially provide economies of scale for the HTL plant.⁴ Swine

manure can be collected within an assumed radius of 80 km. The estimated transportation cost is about \$22/dry tonne swine manure (see previous sections for details) for transporting 25% solids manure 80 km (161 km round trip). Also, an avoided disposal cost is expected in the form of feedstock credits to the HTL plants paid by the farms. A wide range of \$6 - 165/dry tonne is selected as the swine manure management practices vary greatly according to farm operations, farm scale, the type of manure handling system, etc. Based on the estimated transportation and avoided disposal costs, an effective feedstock cost (transportation plus avoided disposal) range of -\$143/dry tonne (credit) to \$17/dry tonne cost is evaluated for the swine manure price at the HTL plant gate. The supporting information provides more details on the estimated transportation and avoided comprehensive nutrient management plans (CNMP) costs. Other economic assumptions investigated in the sensitivity analysis include the uncertainties of HTL and upgrader capital cost, which varied by -40% to +40% of the base case. For the technical variables, variabilities in biocrude yield and hydrotreating fuel oil yield are assessed in this sensitivity study. Biocrude yield varies with the swine manure composition, ash content, and feed solid content. Generally, higher lipid, lower ash, and higher feed solid contents will result in a higher biocrude yield. A wide range of 30-60% is evaluated for the biocrude yield considering the variabilities in swine manure

composition and ash content. Hydrotreating fuel oil yield depends on the heteroatom content such as oxygen, nitrogen, and sulfur in biocrude. A relatively narrow range for hydrotreating yield is selected because HTL-derived biocrude from different feedstocks shows a similar elemental composition and hydrotreating behavior.^{11, 28}

Table S10. Assumptions varied in the sensitivity analysis for the swine manure HTL pathway.

| Process Variables | Optimistic Scenario | Base Case | Conservative Scenario |
|-------------------------------------|------------------------|-----------|--------------------------|
| Economic assumptions: | | | |
| HTL plant scale, dry tonne per day | 227 | 100 | 27 |
| Feedstock price, \$/dry tonne | -143 | 0 | 17 |
| HTL capital cost | -40% | | +40% |
| Upgrader capital cost | -40% | | +40% |
| Technical assumptions: | | | |
| Biocrude yield (dry, ash free), wt% | 60% | 50% | 30% |
| Ash content (dry), wt% | 5% | 12% | 30% |
| HTL feed solid content (dry), wt% | 15% | 25% | 30% |
| Hydrotreating oil yield, wt% | 85% | 81% | 77% |

Material and Energy Balance Results

MCCI Fuel from Yellow Grease to HEFA

Table S11. Material and energy balances, in MJ of MCCI bio-blendstock, of yellow grease hydroprocessing to produce an MCCI bio-blendstock

| Yellow grease to MCCI bio-blendstock | | |
|--------------------------------------|----------|-------|
| Yellow grease | 3.24E-02 | kg/MJ |
| Hydrotreating catalyst | 3.38E-06 | kg/MJ |
| Isomerization/Hydrocracking catalyst | 5.50E-07 | kg/MJ |
| Hydrogen | 4.14E-02 | MJ/MJ |
| Water | 2.02E-02 | L/MJ |
| NG | 8.44E-02 | MJ/MJ |
| Electricity | 1.19E-02 | MJ/MJ |
| C3H8 (Co-product) | 4.77E-02 | MJ/MJ |

MCCI Fuel from Swine Manure HTL

Table S12. Material and energy balances, in MJ of MCCI bio-blendstock, of swine manure HTL to produce biocrude and biocrude upgrading for MCCI bio-blendstock production.

| Swine manure HTL to MCCI bio-blendstock | | |
|--|----------|----------------|
| <i>Swine manure HTL, a 100 dry tonne per day processing capacity</i> | | |
| Sludge | 2.16E+00 | kg/kg biocrude |
| Natural gas | 5.23E+00 | MJ/kg biocrude |
| Electricity (HTL process) | 4.21E-01 | MJ/kgbiocrude |
| Electricity (wastewater treatment plant) | 1.95E+00 | MJ/kgbiocrude |
| Dewatering polymer | 5.67E-03 | kg/kg biocrude |

| | | |
|---|----------|--|
| Quicklime (CaO) | 1.06E-01 | kg/kg biocrude |
| Cooling water makeup | 4.69E-02 | L/kg biocrude |
| <i>Waste treatment and handling, a 100 dry tonne per day processing capacity</i> | | |
| Solid waste handling | | |
| Solids from NH ₃ removal for landfill (85% CaCO ₃) | 2.19E-01 | kg/kg biocrude |
| CO ₂ emission factor of landfilled solids from NH ₃ removal | 2.16E-01 | g CO ₂ /g CaCO ₃ |
| HTL solids for landfill (58% moisture) | 8.57E-01 | kg/kg biocrude |
| Carbon content in HTL solids | 1.31E+01 | wt. % |
| Carbon sequestration ratio for HTL solids | 8.00E+01 | wt. % |
| Aqueous waste treatment | | |
| CHG catalyst for aqueous waste treatment ^a | 2.36E-03 | kg/kg biocrude |
| <i>Biocrude upgrading to renewable diesel, a 144 million liters per year production capacity</i> | | |
| Biocrude | 3.07E-02 | kg/MJ of fuel |
| Natural gas for process heating | 4.09E-02 | MJ/MJ of fuel |
| Natural gas for H ₂ production | 1.29E-01 | MJ/MJ of fuel |
| Electricity | 9.16E-03 | MJ/MJ of fuel |
| Cooling tower chemical | 1.35E-07 | kg/MJ of fuel |
| Boiler chemical | 2.06E-07 | kg/MJ of fuel |
| Hydrotreating catalyst (CoMo/ γ -Al ₂ O ₃) | 5.98E-06 | kg/MJ of fuel |
| Hydrotreating catalyst (NiMo/ γ -Al ₂ O ₃) | 2.58E-06 | kg/MJ of fuel |
| Hydrocracking catalyst | 3.89E-07 | kg/MJ of fuel |
| Hydrogen plant catalyst | 3.11E-07 | kg/MJ of fuel |
| Cooling water makeup | 1.53E-02 | L/MJ of fuel |
| Boiler feedwater makeup | 7.50E-03 | L/MJ of fuel |

^aHydrotreating catalyst is used as a surrogate for the ruthenium catalyst used in CHG due to lack

of data for ruthenium catalyst in GREET.

MFSP of the Biocrude from Swine Manure HTL

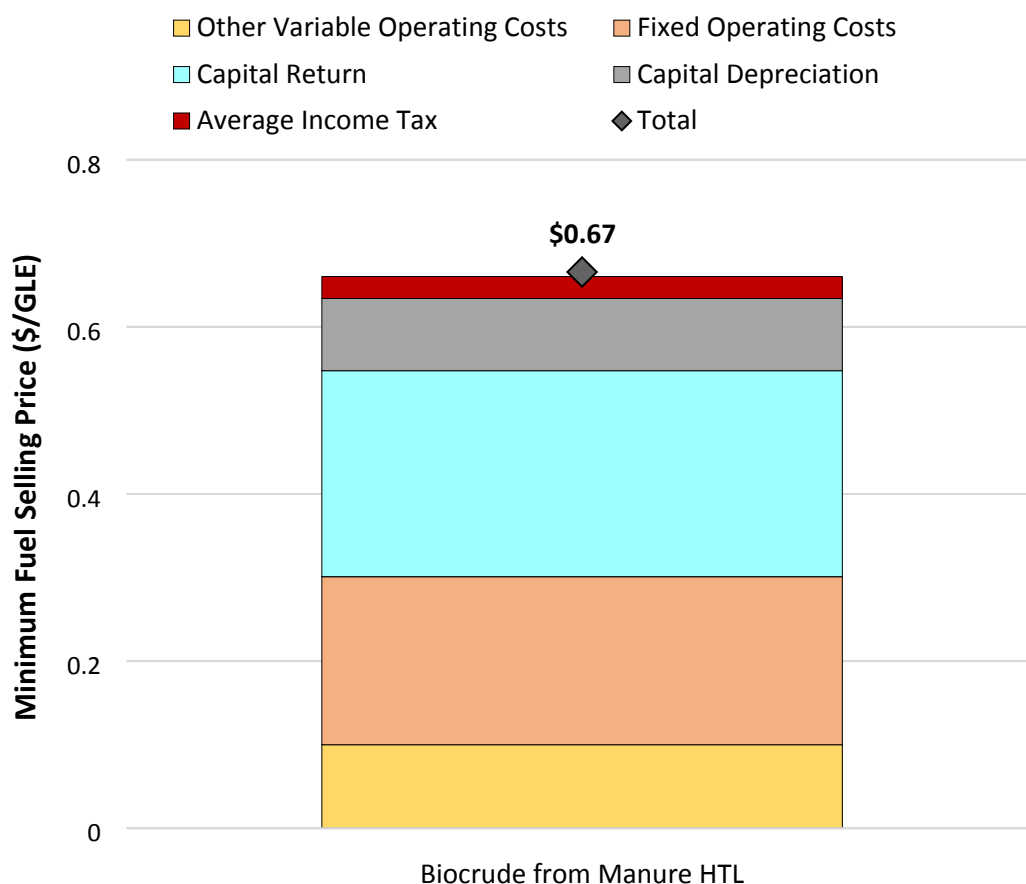


Fig. S4. Breakdown of the MFSP of the biocrude from swine manure HTL.

Life-Cycle Water Consumption, Fossil Energy Consumption, NO_x Emissions, and PM_{2.5} Emissions Results

Fig. S5 shows the life-cycle water consumption of MCCI fuels produced with the yellow grease to HEFA and the manure HTL pathways. The yellow grease to HEFA pathway achieves 60%

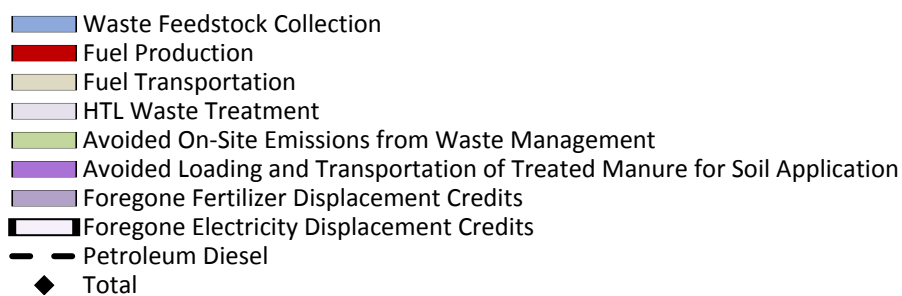
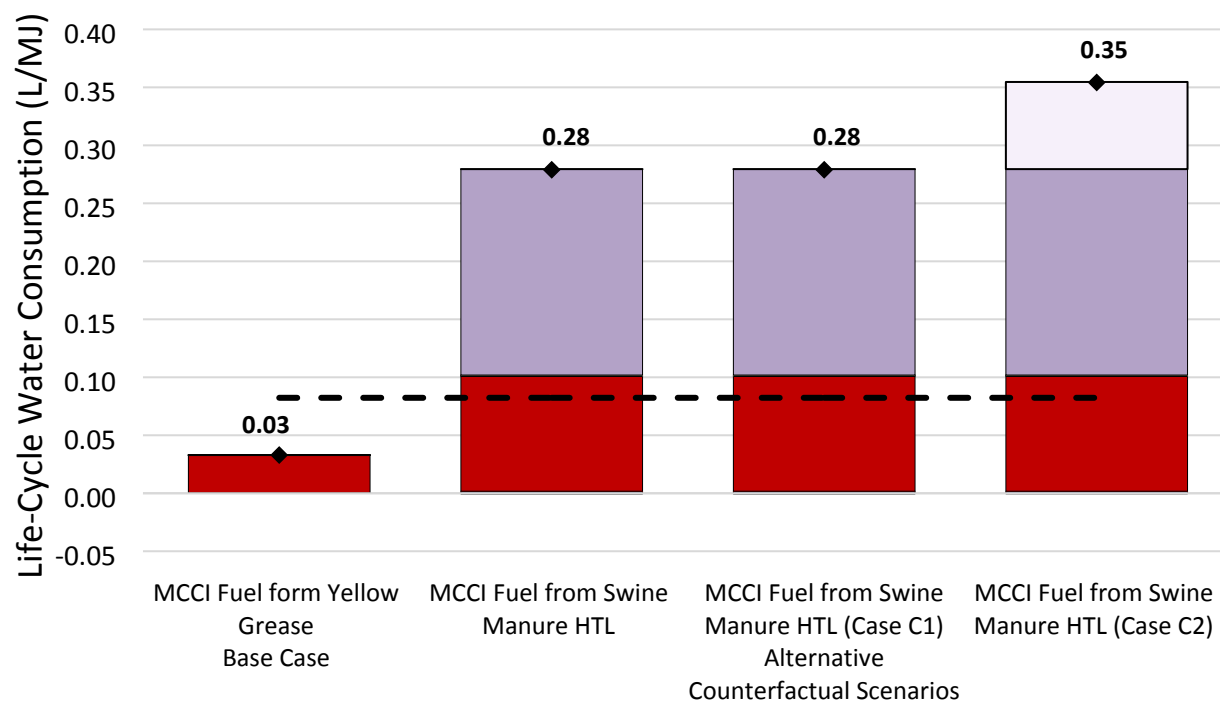
lower life-cycle water usage than petroleum diesel. The life-cycle water usage is mainly driven by the water used for the conversion of yellow grease to RD. On the other hand, the manure HTL pathway may cause an over 200% increase in water consumption relative to petroleum diesel. There are two major contributors to water usage. The first is the forgone credits from the production of the synthetic fertilizers that are displaced in the counterfactual scenario;²⁹⁻³⁰ the second is the upstream water usage for the natural gas and electricity used during HTL and upgrading. If the manure is from a farm with dedicated anaerobic digesters, the foregone credits from displacing grid electricity would further increase the life-cycle water usage for manure HTL.

Both the yellow grease and manure pathways consume less life-cycle fossil energy than petroleum diesel. Like water usage, fuel production is an important contributor to fossil energy consumption for both pathways. For manure HTL, the foregone synthetic fertilizer displacement credits are also significant. If the manure is from a farm with dedicated anaerobic digesters, the foregone credits from electricity displacement would drive the life-cycle fossil energy further up.

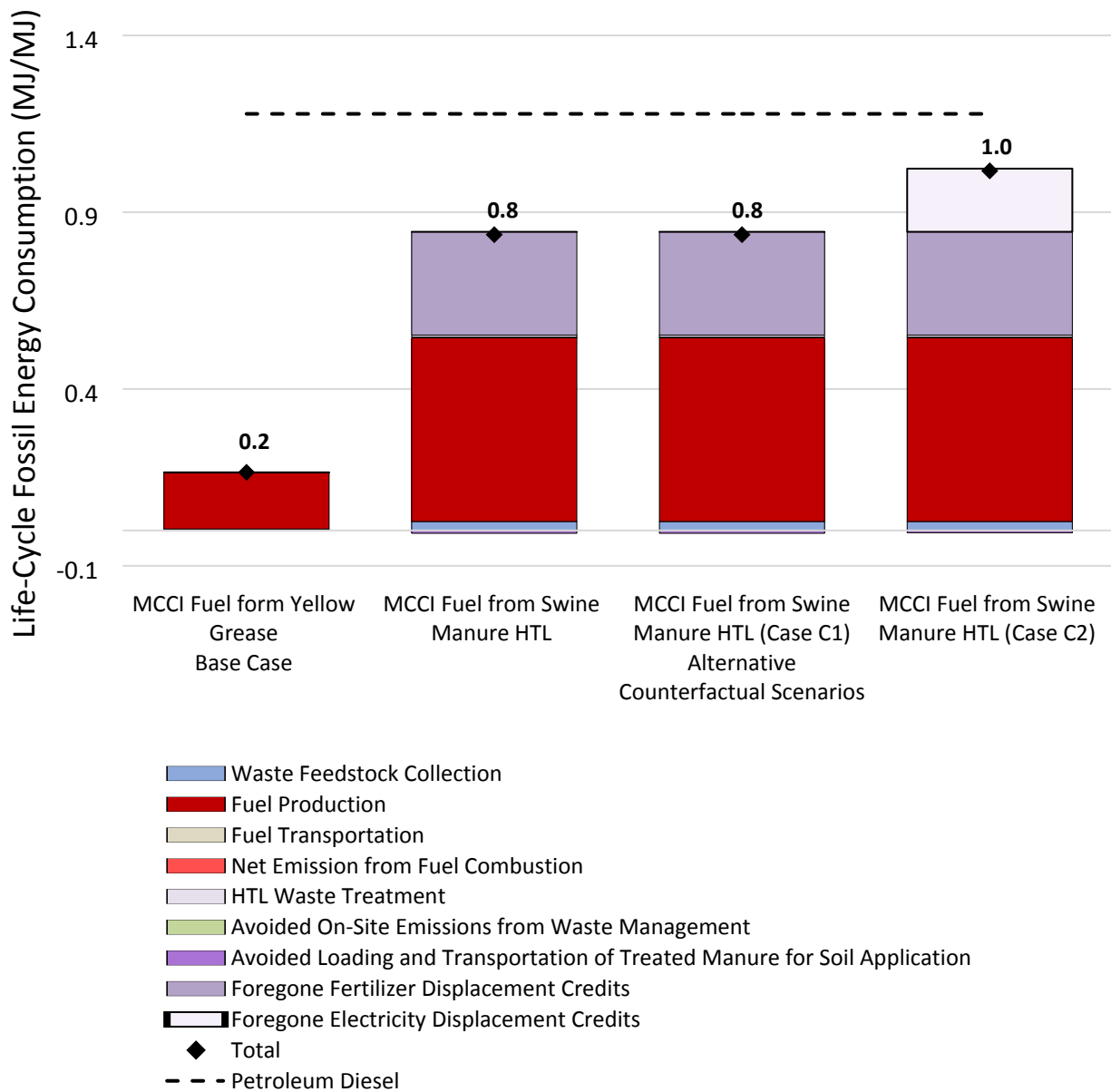
The life-cycle NO_x emissions are mainly driven by three contributors: fuel combustion in vehicles, fuel production, and foregone fertilizer displacement credits (for manure HTL only). The yellow grease to HEFA pathway achieves lower life-cycle NO_x emissions than petroleum diesel,

while the manure HTL pathway generates higher NO_x emissions. It is also noted that different counterfactual scenarios do not affect the NO_x emission greatly, because the on-site combustion of biogas in CHP generates similar NO_x emission to that from the U.S. average electricity generation mix if manure is used for AD and electricity generation in the counterfactual scenario.

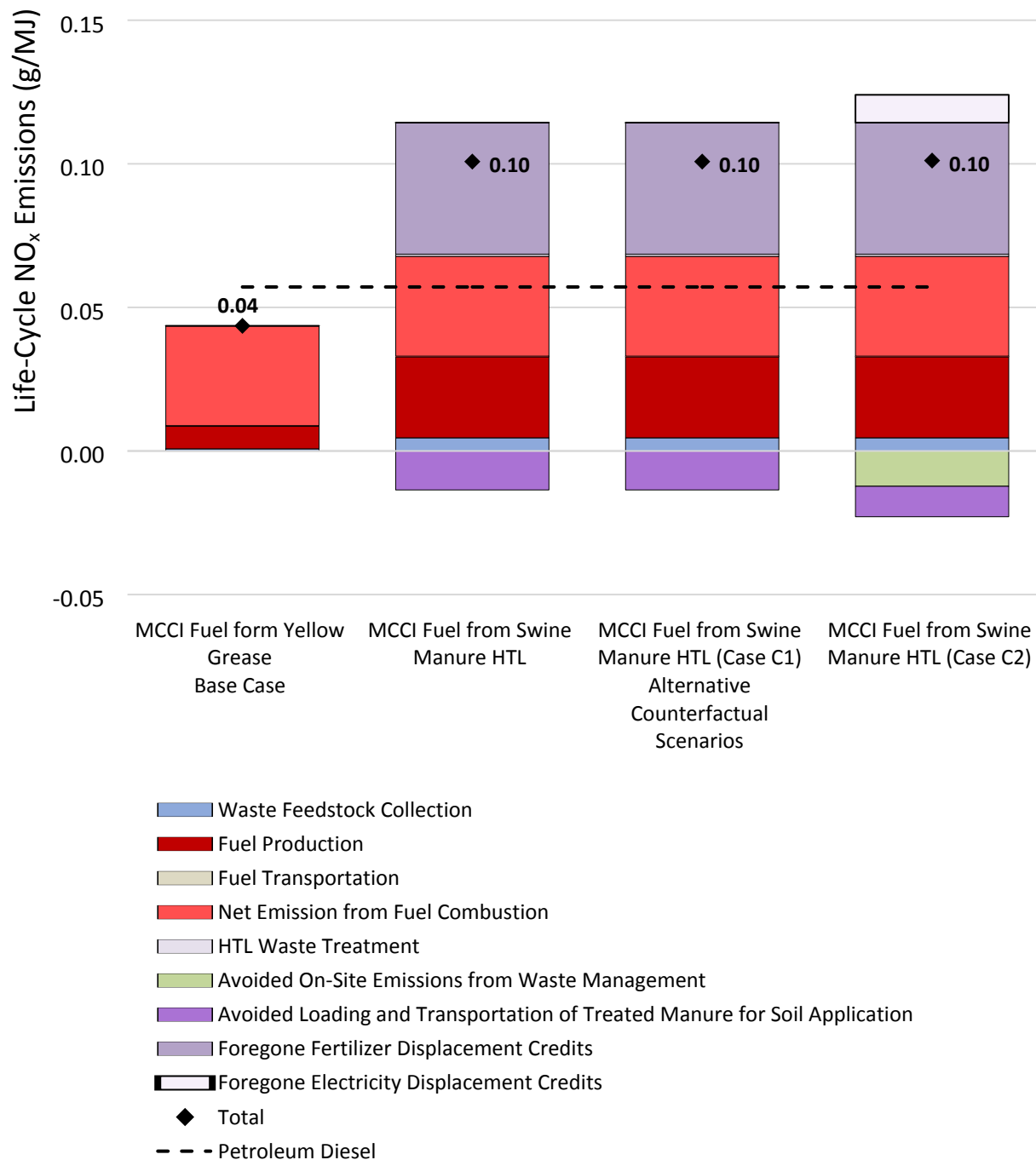
Particulate matter (PM) emission presents a similar trend as NO_x emissions. The yellow grease to HEFA pathway causes slightly less PM emission than petroleum diesel. However, the manure HTL pathway has much higher PM emission than petroleum diesel due to the foregone fertilizer displacement credits in the counterfactual scenario. The impact of various counterfactual scenarios on the manure HTL pathway is small.



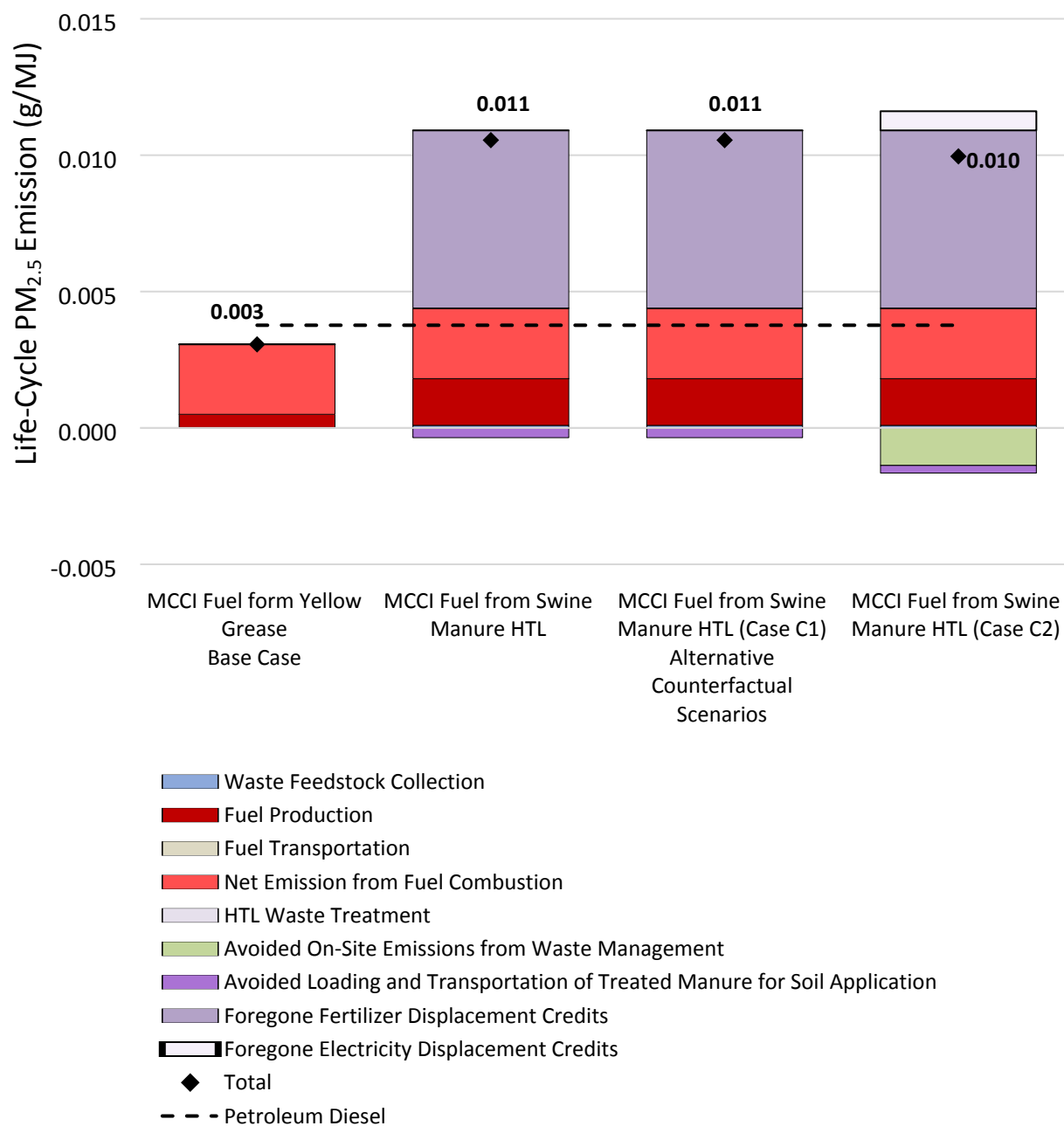
(a)



(b)



(c)



(d)

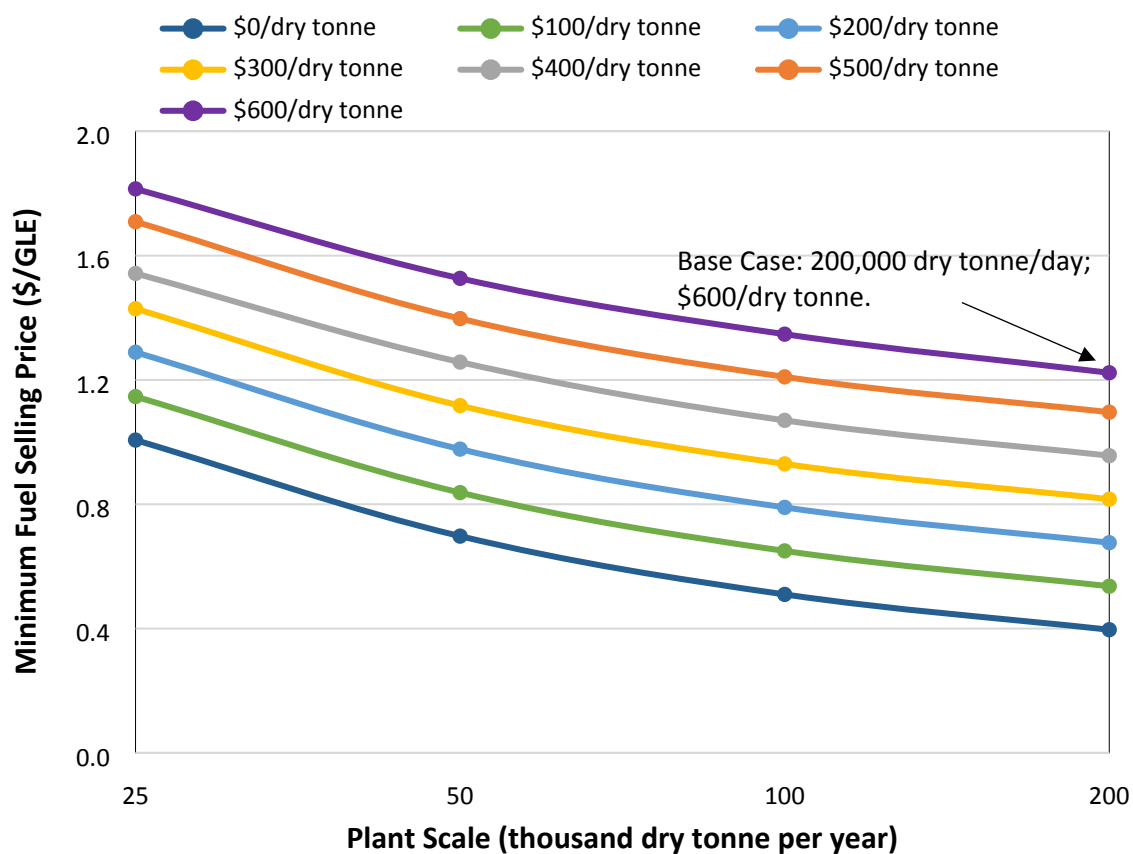
Fig. S5. Life-cycle (a) water consumption, (b) fossil energy consumption, (c) NO_x emission, and (d) PM_{2.5} emission results for MCCI bio-blendstocks produced with yellow grease via

hydroprocessing and swine manure via hydrothermal liquefaction (HTL). Three counterfactual scenarios were considered for the swine manure HTL pathway: Base Case assumes average U.S. swine manure management practices with 50% of the manageable biogas flared and the remaining vented; Case C1 assumes average U.S. swine manure management practices with 100% of the manageable biogas flared; Case C2 assumes anaerobic digestion for bioelectricity generation in the counterfactual scenario. No counterfactual scenario is considered for MCCI bio-blendstocks from yellow grease because yellow grease has been commoditized and its price is determined by market demand.

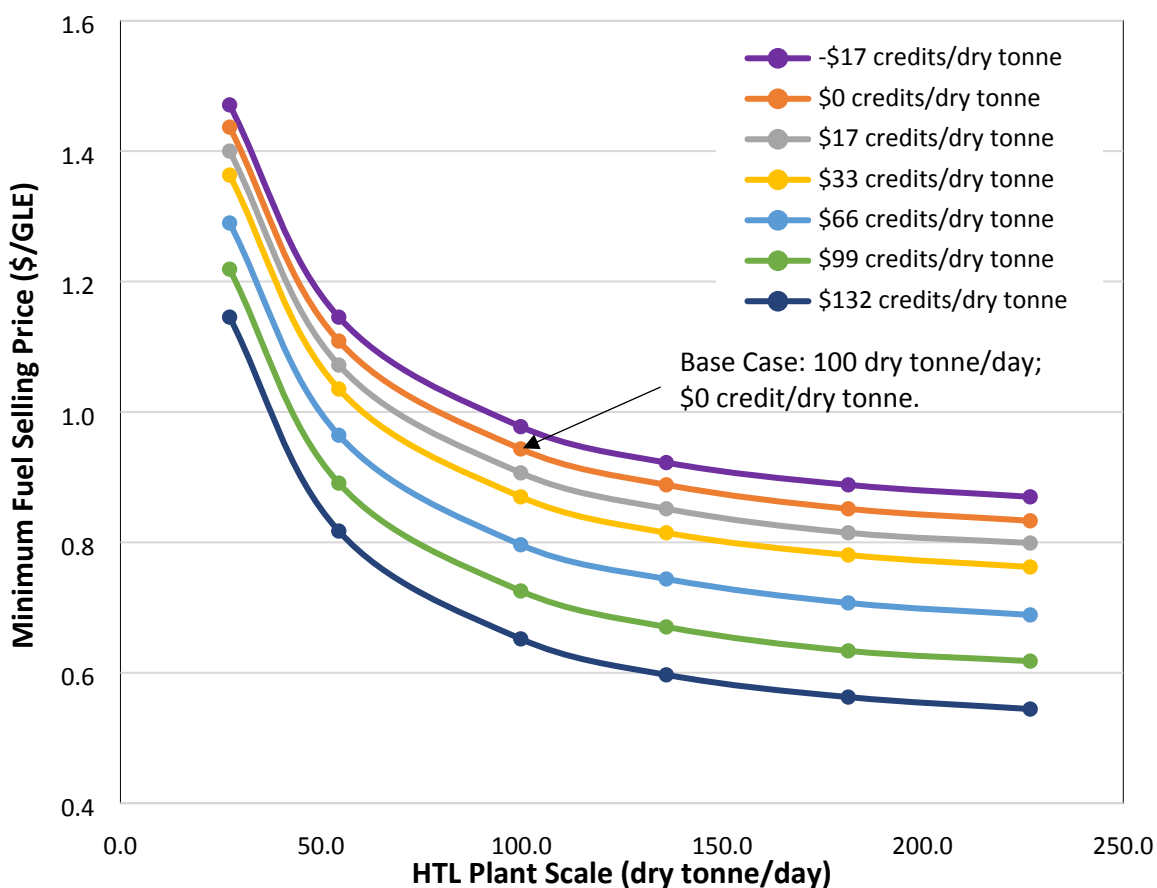
Process Scalability

A key challenge of waste-to-fuel scalability is related to the viability of individual waste-to-fuel resource characteristics, including their location, amount, and quality. The distributed waste resources are a well-known feature for any conversion strategy using a waste resource. A total of 5.4 million dry tonnes of FOG is available in the U.S.² Animal fats contribute more than 50% of the total resource, brown grease contributes about 28%, and yellow grease about 19% of total FOG generation.² To improve the intrinsic scalability issues facing high-cost HEFA fuel, the authors recommend that increasing plant scale by centralizing plant oil collection stations and blending various plant oils with waste oils (such as yellow grease) must be realized to make MFSP

approaching \$0.7/GLE or lower. A sensitivity analysis is illustrated in Figure S6(a) on both plant scales and yellow grease costs.



(a)



(b)

Figure S6. Effect of plant scale and feedstock costs/credits (avoided disposal cost) for MCCI fuel produced from (a) yellow grease and (b) swine manure HTL.

The increased demand for animal fats and used cooking oil in biodiesel and renewable diesel production is being driven by the LCFS. The LCFS credits would facilitate the deployment of the technology in the early stage and have measurable GHG impacts in the long term. Under this

standard, these products are preferred due to their low carbon intensity (CI) scores over other feedstocks. Used cooking oil has some of the lowest CI scores, followed by distiller's corn oil, animal fats, and finally vegetable oils.³¹

The scale of the processing plant is the most important cost driver for the HTL plant due to manufacturing economies of scale. To further analyze the impacts of plant scales and feedstock credits, sensitivity analyses were conducted by varying plant scales from 27-227 dry tonne/day and feedstocks from -\$17 to \$132/dry tonne. As shown in Figure S6(b), combining large-scale and feedstock credits could decrease the MFSP and make this process more attractive. On a scale of half the base case (50 dry tonne/day; 0 feedstock credit), the MFSP increased by 21%. On twice the scale of the base case (200 dry tonne/day; 0 feedstock credit), MFSP was reduced to \$0.84/GLE. For the base case scale (100 dry tonne/day), receiving feedstock credits of \$132/dry tonne decreased the MFSP by \$1.10/GGE while adding \$17/dry tonne feedstock costs increased the MFSP to \$0.98/GLE.

A key issue for consideration of scalability of renewable diesel production from manure HTL is feedstock delivery. Manure supply from existing manure farms ranges from <0.5 to >45 dry tonne per day (Figure S2). Large farms (>9 dry tonne per day) only account for 0.5% of the total available

swine manure in the United States. A commercial HTL facility is likely to source manure feedstock from multiple swine farms. An efficient feedstock supply chain is important for commercial-scale production of biofuels from high-moisture feedstocks such as animal manure because the cost of feedstock transportation is high. Regions with high availability of livestock manure such as the Midwest and Great Lakes region are thus more suitable for large-scale manure HTL. Another potential way to alleviate the issue is to blend swine manure with other wet feedstocks such as wastewater sludge and food waste.³²

The designed manure HTL process requires a centralized upgrading facility to take advantage of economies of scale. It is thus important for the siting of the centralized upgrading facility to minimize the cost and time of transporting the biocrude from the HTL plant to the upgrading facility. The design of a system for minimized biocrude degradation and emission of volatile compounds during its storage and transportation is also important.³³

References

1. Snowden-Swan, L. J.; Zhu, Y.; Bearden, M. D.; Seiple, T. E.; Jones, S. B.; Schmidt, A. J.; Billing, J. M.; Hallen, R. T.; Hart, T. R.; Liu, J.; Albrecht, K. O.; Fox, S. P.; Maupin, G. D.; Elliott, D. C. *Conceptual Biorefinery Design and Research Targeted for 2022: Hydrothermal*

- Liquefaction Processing of Wet Waste to Fuels*; PNNL-27186; Pacific Northwest National Lab. (PNNL): Richland, WA (United States), 2017. DOI 10.2172/1415710
2. Milbrandt, A.; Seiple, T.; Heimiller, D.; Skaggs, R.; Coleman, A. Wet waste-to-energy resources in the United States. *Resour., Conserv. Recycl.* **2018**, *137*, 32-47, DOI 10.1016/j.resconrec.2018.05.023
 3. Marufuzzaman, M.; Ekşioğlu, S. D.; Hernandez, R. Truck versus pipeline transportation cost analysis of wastewater sludge. *Transp. Res. Part A: Policy Pract.* **2015**, *74*, 14-30, DOI 10.1016/j.tra.2015.02.001
 4. United States Department of Agriculture *Costs Associated with Development and Implementation of Comprehensive Nutrient Management Plans*; 2003.
 5. Pearlson, M. N. A techno-economic and environmental assessment of hydroprocessed renewable distillate fuels. Massachusetts Institute of Technology, 2011.
 6. NESTE Neste MY Renewable Diesel – high-performing low-carbon biofuel. <https://www.neste.com/products/all-products/renewable-road-transport/neste-my-renewable-diesel#832ce7c2> (accessed April 19, 2021).
 7. Honeywell UOP Honeywell Introduces Simplified Technology to Produce Renewable Diesel <https://uop.honeywell.com/en/news-events/2021/january/honeywell-uop-ecofining-single-stage-process> (accessed April 19, 2021).
 8. Chemicals Technology Dynamic Fuels Renewable Synthetic Fuel Plant. <https://www.chemicals-technology.com/projects/dynamicfuelslouisian/#:~:text=The%20Dynamic%20Fuels%20plant%20is,fuel%20plant%20in%20North%20America> (accessed April 19, 2021).
 9. Diamond Green Diesel A Renewable Fuel For a Low-Carbon World. <https://www.diamondgreendiesel.com/> (accessed April 19, 2021).
 10. AspenTech Aspen Plus. Release 10.0. (accessed May 17, 2021).
 11. Snowden-Swan, L. J.; Billing, J. M.; Thorson, M. R.; Schmidt, A. J.; Santosa, D. M.; Jones, S. B.; Hallen, R. T. *Wet Waste Hydrothermal Liquefaction and Biocrude Upgrading to Hydrocarbon Fuels: 2019 State of Technology*; PNNL-29882; Pacific Northwest National Lab. (PNNL): Richland, WA (United States), 2020. DOI 10.2172/1617028
 12. U.S. Environmental Protection Agency *Inventory of US Greenhouse Gas Emissions and Sinks: 1990–2018*; EPA 430-R-20-002; 2020.
 13. Jones, S. B.; Zhu, Y.; Anderson, D. B.; Hallen, R. T.; Elliott, D. C.; Schmidt, A. J.; Albrecht, K. O.; Hart, T. R.; Butcher, M. G.; Drennan, C.; Snowden-Swan, L. J.; Davis, R.; Kinchin, C. *Process Design and Economics for the Conversion of Algal Biomass to Hydrocarbons: Whole Algae Hydrothermal Liquefaction and Upgrading*; PNNL-23227; Pacific Northwest National Lab. (PNNL): Richland, WA (United States), 2014. DOI 10.2172/1126336
 14. Wang, Z.; Dunn, J. B.; Han, J.; Wang, M. Q. Effects of co-produced biochar on life cycle greenhouse gas emissions of pyrolysis-derived renewable fuels. *Biofuels, Bioprod. Biorefin.* **2014**, *8* (2), 189-204, DOI 10.1002/bbb.1447
 15. Cai, H.; Wang, M.; Han, J. *Update of the CO₂ Emission Factor from Agricultural Liming*; 2014.
 16. Chastain, J. P. Fertilizer Value of Swine Manure: A Comparison of a Lagoon and a Deep Pit Slurry System. <https://lpelc.org/fertilizer-value-of-swine-manure-a-comparison-of-a-lagoon-and-a-deep-pit-slurry-system/> (accessed March 15, 2021).
 17. U.S. Environmental Protection Agency *Annexes to the Inventory of U.S. GHG Emissions and Sinks*; 2020.

18. U.S. Environmental Protection Agency *Market Opportunities for Biogas Recovery Systems at U.S. Livestock Facilities*; EPA-430-R-18-006; 2018.
19. U.S. Environmental Protection Agency Livestock Anaerobic Digester Database. <https://www.epa.gov/agstar/livestock-anaerobic-digester-database> (accessed March 15, 2021).
20. Eggleston, H.; Buendia, L.; Miwa, K.; Ngara, T.; Tanabe, K. *2006 IPCC Guidelines for National Greenhouse Gas Inventories*. IPCC: 2006.
21. Bruun, S.; Hansen, T. L.; Christensen, T. H.; Magid, J.; Jensen, L. S. Application of processed organic municipal solid waste on agricultural land – a scenario analysis. *Environ. Model. Assess.* **2006**, *11* (3), 251-265, DOI 10.1007/s10666-005-9028-0
22. Nguyen, T. L. T.; Hermansen, J. E.; Mogensen, L. Fossil energy and GHG saving potentials of pig farming in the EU. *Energy Policy* **2010**, *38* (5), 2561-2571, DOI 10.1016/j.enpol.2009.12.051
23. Aguirre-Villegas, H. A.; Larson, R.; Reinemann, D. J. From waste-to-worth: energy, emissions, and nutrient implications of manure processing pathways. *Biofuels, Bioprod. Biorefin.* **2014**, *8* (6), 770-793, DOI 10.1002/bbb.1496
24. Wiens, M. J.; Entz, M. H.; Wilson, C.; Ominski, K. H. Energy requirements for transport and surface application of liquid pig manure in Manitoba, Canada. *Agric. Syst.* **2008**, *98* (2), 74-81, DOI 10.1016/j.agry.2008.03.008
25. Han, J.; Mintz, M.; Wang, M. *Waste-to-wheel analysis of anaerobic-digestion-based renewable natural gas pathways with the GREET model*; Argonne National Lab.(ANL), Argonne, IL (United States): 2011.
26. Mintz, M.; Han, J.; Wang, M.; Saricks, C. *Well-to-Wheels analysis of landfill gas-based pathways and their addition to the GREET model*; ANL/ESD/10-3; Argonne National Lab.(ANL): Lemont IL (United States), 2010.
27. SRECTrade LCFS Market Update – February 2021. <https://www.srectrade.com/blog/srec-markets/lcfs-market-update-february-2021> (accessed March 29, 2021).
28. Elliott, D. C.; Biller, P.; Ross, A. B.; Schmidt, A. J.; Jones, S. B. Hydrothermal liquefaction of biomass: Developments from batch to continuous process. *Bioresour. Technol.* **2015**, *178*, 147-156, DOI 10.1016/j.biortech.2014.09.132
29. Lampert, D. J.; Cai, H.; Elgowainy, A. Wells to wheels: water consumption for transportation fuels in the United States. *Energy Environ. Sci.* **2016**, *9* (3), 787-802, DOI 10.1039/C5EE03254G
30. Johnson, M. C.; Palou-Rivera, I.; Frank, E. D. Energy consumption during the manufacture of nutrients for algae cultivation. *Algal Res.* **2013**, *2* (4), 426-436, DOI 10.1016/j.algal.2013.08.003
31. California Air Resources Board LCFS Pathway Certified Carbon Intensities. <https://ww2.arb.ca.gov/resources/documents/lcfs-pathway-certified-carbon-intensities> (accessed May 17, 2021).
32. Skaggs, R. L.; Coleman, A. M.; Seiple, T. E.; Milbrandt, A. R. Waste-to-Energy biofuel production potential for selected feedstocks in the conterminous United States. *Renewable Sustainable Energy Rev.* **2018**, *82*, 2640-2651, DOI 10.1016/j.rser.2017.09.107
33. Palomino, A.; Godoy-Silva, R. D.; Raikova, S.; Chuck, C. J. The storage stability of biocrude obtained by the hydrothermal liquefaction of microalgae. *Renewable Energy* **2020**, *145*, 1720-1729, DOI 10.1016/j.renene.2019.07.084