

# **Integrating Anaerobic Digestion, Hydrothermal Liquefaction, and Biomethanation within a Power-to-Gas framework for dairy waste management and grid decarbonization: a techno-economic analysis.**

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## **Supplemental Information**

### **1. Bioenergetics to determine AD biogas CO<sub>2</sub>/CH<sub>4</sub> ratio**

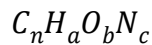
Using bioenergetics and assuming BOD is represented by a composite VFA (composite of the three main VFAs; butyrate, propionate and acetate), the stoichiometric equation for anaerobic conversion of the BOD into methane and carbon dioxide is derived to compute the biogas CO<sub>2</sub>/CH<sub>4</sub> volumetric ratio.

Conversion of complex or organic matter into methane starts with the hydrolysis of carbohydrates, fats, and proteins into amino acids and fatty acids. These acids are then utilized for their energy for growth by fermenting bacteria. This produces organic acids and hydrogen, which are the main substrates used by methanogenic archaea, which convert them into methane.

The process is broken down into two simple steps; hydrolysis and methane formation. The complex organics are hydrolyzed to form the volatile fatty acids. There has to be a balance between the hydrolytic and fermentative bacteria, achieved through managing the pH of the reactor. The main Volatile organic compounds (VOC) formed as a result are acetic acid, propionic acid, and butyric acid.

Mass balances on carbon, nitrogen, oxygen and hydrogen are maintained throughout the reaction, where the most important balance is the electron balance. The removal of BODL depends totally on the methane formation. If nitrogen and sulfur are used in the reaction, these are usually present in the products as sulfides and ammonium.

The empirical molecular formula of an electron donor to be:



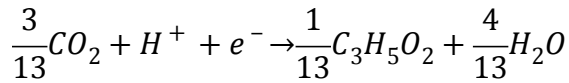
Where a portion of the electron equivalence is synthesized to biomass and nitrogen. CO<sub>2</sub> is not the true electron acceptor. The value  $f_s^0$  represents the fraction of waste organic matter synthesized or converted to cells, while  $f_e$  represents the portion of organic matter converted to energy, therefore:

$$f_s^0 + f_e = 1$$

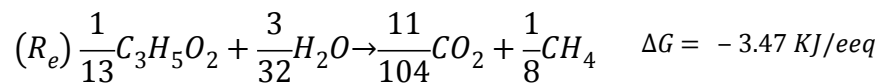
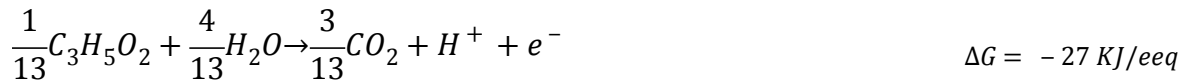
The  $f_s$  depends on the decay rate, residence time, cell energy generation and synthesis reactions. It can be calculated using:

$$f_s = f_s^0 \left[ \frac{1 + (1 - f_d)b\theta_x}{1 + b\theta_x} \right]$$

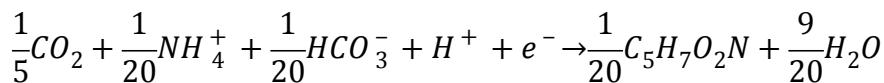
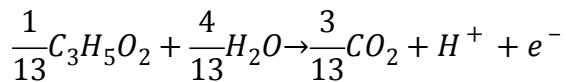
Where b is the decay rate, and  $\theta_x$  is the mean residence time. Taking a composite of the three main VFA's in manure, acetate, butyrate and lactate, a composite formula,  $C_3H_5O_2$ , is derived. Using the generalized half reaction equation, the ion half equation is determined:



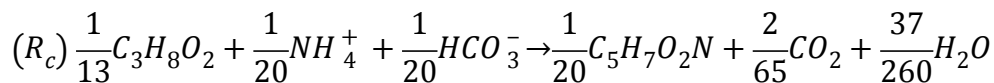
This reaction has a Gibbs Free Energy of -27 KJ/eeq, this is the amount of energy required to convert the composite VFA to carbon dioxide. This along with the half reaction for methane can be used to determine the reaction equation for energy and the reaction for cells.



Then the cell synthesis reaction is given by:



Therefore,



Using the relationship of the Gibbs Free Energy, we can calculate the number of electrons going to energy production, A. The reaction proceeds metabolically via pyruvate from the initial VFA's, and then this is converted to cell carbon.

$$\Delta G_P = \Delta G_{pyruvate}^{o'} - \Delta G_{VFA\ Composite}^{o'} = 35.09 - 27.0 = 8.09 \text{ KJ/eeq}$$

The energy needed to convert pyruvate to cell carbon is empirical and is given to be  $\Delta G_C = 18.8 \text{ KJ/eeq}$ . Using the relationship between the Gibbs free energy of reaction and of carbon and pyruvate, the number of electrons are calculated.

$$A = \frac{-\left(\frac{\Delta G_P}{\varepsilon^n} + \frac{\Delta G_C}{\varepsilon}\right)}{\varepsilon \Delta G_r} = \frac{-\left(\frac{8.09}{0.6} + \frac{18.8}{0.6}\right)}{0.6 \times 3.47} = 21.5 \frac{\text{eeq energy}}{\text{eeq synthesis}}$$

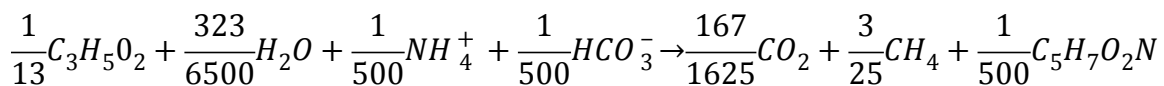
Therefore, the fraction of electron donors going to energy and cell synthesis respectively are given by:

$$f_e^o = \frac{A}{1 + A} = 0.96, \quad f_s^o = \frac{1}{1 + A} = 0.04$$

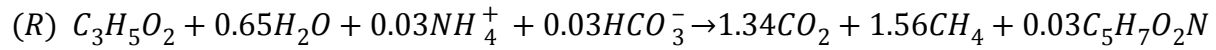
The overall reaction can then be calculated from the following equation:

$$R = f_e^o R_e + f_s^o R_C$$

Combining the two equations used previously, the following overall equation is derived:



This can be further simplified to represent one mole of composite manure:



Therefore, this represents a volume percentage of 56% methane and 44% carbon dioxide, derived from the stichometry. (46% CO<sub>2</sub> and 54% methane molar based)

## 2. Mass balances

Raw manure consists of an aqueous mixture of carbohydrates, protein, lipids, minerals and nutrients<sup>1</sup>. The amount of volatile solids (VS) produced daily by a dairy lactating cow is 7.5 kg<sup>2</sup>, and represents the organic material present in the manure slurry. As such, the total amount of VS fed into a digester is given by:

$$VS (kg) = 7.5 \times cow\#$$

Due to the complexity and variability of the manure feedstock composition, the following set of assumptions were established: the VS consists mainly of soluble volatile fatty acids (VFA) and particulate lignocellulosic material. For simplicity, we assumed lignocellulose to be represented by cellulose (C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>)<sub>n</sub>, while the VFA were represented by a composite chemical formula – assuming equal distribution of acetate, butyrate, propionate – (C<sub>3</sub>H<sub>5</sub>O<sub>2</sub>). Finally, we assumed a VFA/cellulose ratio in VS of 10%/90% consistent with the literature<sup>1</sup>.

The input AD carbon masses (kg/day) were computed as follow:

$$Carbon_{VFA} = \frac{0.10 \times VS}{73 \frac{g \ VFA}{mol \ VFA}} \times 3 \frac{mol \ C}{mol \ VFA} \times 12 \frac{gC}{molC}$$

$$Carbon_{cellulose} = \frac{0.90 \times VS}{162 \frac{g \text{ cellulose}}{mol \text{ cellulose}}} \times 6 \frac{mol \text{ C}}{mol \text{ cellulose}} \times 12 \frac{gC}{molC}$$

Methane generation from the anaerobic digestion of dairy manure was calculated using a methane yield of 264 L CH<sub>4</sub>/kg VS fed, obtained from the literature<sup>3-5</sup>. The carbon mass of CH<sub>4</sub> was calculated as follows:

$$Carbon_{CH_4} = \frac{\left[ methane \left( \frac{L}{d} \right) \times 10^{-3} \frac{m^3}{L} \times \rho_{CH_4} \left( \frac{kg}{m^3} \right) \right]}{16 \frac{g \text{ CH}_4}{mol \text{ CH}_4}} \times 12 \frac{gC}{molC}$$

Where  $methane = VS \times 264$  and  $\rho_{CH_4}$  is the methane density at NTP conditions, 0.668 kg/m<sup>3</sup>. The mass of carbon in CO<sub>2</sub> is obtained similarly after using the CH<sub>4</sub>:CO<sub>2</sub> ratio computed in section S1. Finally, the total mass of carbon in the digestate is computed by subtracting the carbon mass in the biogas (CH<sub>4</sub> and CO<sub>2</sub>) from the total carbon mass in the input. Total carbon mass in the digestate is 0.213 kg C/kg VS/day.

Knowing the total amount of carbon in the digestate, the mass of carbon in the different HTL products can be computed using carbon conversion yields obtained from Posmanik et al. (2017)<sup>6</sup>. The mass of the different HTL product fractions can be calculated by knowing the chemical composition of each fraction. The biocrude oil is rich in carbon and hydrogen and has minimal amount of oxygen and nitrogen<sup>6</sup>. The elemental composition of the biocrude fraction is 73%, 8%, 15.8% and 3.3% carbon, hydrogen, oxygen and nitrogen by weight, respectively (C<sub>28</sub>H<sub>34</sub>O<sub>4</sub>N). The aqueous phase is composed primarily of lactic acid (C<sub>3</sub>O<sub>3</sub>H<sub>5</sub>) and acetic acid (C<sub>2</sub>O<sub>2</sub>H<sub>4</sub>), with a 40/60 percentage distribution respectively<sup>6</sup>. The solid phase, i.e., hydro-char is primarily composed of lignin<sup>6</sup>, (C<sub>81</sub>H<sub>92</sub>O<sub>28</sub>)<sup>7</sup>. Table S1 shows the HTL carbon and mass balances.

**Table S1 – HTL products characteristics and mass balance (per 1000 kg C input in digestate)**

HTL product	Chemical	Carbon	Conversion yield	Mass of	Mass of HTL
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	Formula	fraction (g C/ g product)	(g C product/ g C feed) <sup>6</sup>	carbon (kg)	product (Kg) <sup>a</sup>
<b>Biocrude oil</b>	C <sub>28</sub> H <sub>34</sub> O <sub>4</sub> N	0.73	0.38	380	520
<b>Hydro-char</b>	C <sub>81</sub> H <sub>92</sub> O <sub>28</sub>	0.64	0.24	240	375
<b>Aqueous phase</b>			0.19	190	
<b>Acetic acid</b>	C <sub>2</sub> O <sub>2</sub> H <sub>4</sub>	0.40		114	285
<b>(60%)</b>	C <sub>3</sub> O <sub>3</sub> H <sub>5</sub>	0.40		76	190
<b>Lactic Acid</b>					
<b>(40%)</b>					
<b>Gas</b>	CO <sub>2</sub>	0.27	0.19	190	704

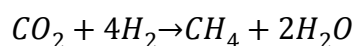
**a Carbon mass/carbon fraction of product**

### 3. Electrolysis Efficiency

The electrolysis efficiency assumed in this study is 70% (HHV). This was calculated based on Electrochaea's reported overall power-to-gas efficiency of 50 m<sup>3</sup> CO<sub>2</sub>/h conversion per 1 MW<sub>e</sub> of electrolyzer capacity, such that a 1 MW<sub>e</sub> P2G size produces enough hydrogen to convert 50 Nm<sup>3</sup> CO<sub>2</sub>/h into CH<sub>4</sub>.

$$1 \text{ MW}_e \rightarrow 50 \frac{\text{m}^3 \text{CO}_2}{\text{h}}$$

In this section, we show how the electrolysis efficiency is calculated. Electrochaea's BioCat methanation reaction, reported to have a 100% efficiency<sup>8</sup>, is shown in the equation below:



Equation shows that every mole of CO<sub>2</sub> reacts with four moles of H<sub>2</sub>. For a 1MW<sub>e</sub> electrolyzer, the mass of hydrogen can then be calculated by:

$$\begin{aligned} 50 \frac{\text{m}^3 \text{CO}_2}{\text{h}} \times 1.98 \frac{\text{kg CO}_2}{\text{m}^3 \text{CO}_2} \times 10^3 \frac{\text{g}}{\text{kg}} \times \frac{1 \text{ mol CO}_2}{44 \text{ g CO}_2} \times 4 \frac{\text{mol H}_2}{\text{mol CO}_2} \times 2 \frac{\text{g H}_2}{\text{mol H}_2} \times 10^{-3} \frac{\text{kg}}{\text{g}} \\ = 18 \frac{\text{kg H}_2}{\text{h}} \end{aligned}$$

So, a 1MW<sub>e</sub> electrolyzer produces 18 kg H<sub>2</sub>/h. Using hydrogen's higher heating value (HHV<sub>H<sub>2</sub></sub> = 141 MJ/kg), the embedded thermal energy is calculated to be 2538 MJ/h. The electrolyzer efficiency (HHV based) is then calculated by:

$$\begin{aligned} 1 \text{ MW}_e \rightarrow 2538 \frac{\text{MJ}}{\text{h}} \\ 1 \text{ MW}_e \text{h} \rightarrow 2538 \text{ MJ} \end{aligned}$$

$$3600 \text{ MJ}_e \rightarrow 2538 \text{ MJ}$$

$$\eta_{HHV} = \frac{2538}{3600} \approx 70\%$$

Using the lower heating value ( $\text{LHV}_{\text{H}_2} = 121 \text{ MJ/kg}$ ), we obtain an efficiency of 60%.

#### 4. LCFS and RFS renewable fuel credit calculation

RNG qualifies for sale under both the EPA's Renewable Fuel Standard (RFS) and under California's Low Carbon Fuel Standard (LCFS). This section details the methods used in calculating the renewable fuel credits.

##### RFS

Fuels under the RFS are assigned a D-code (based on EPA pre-approved pathways), representing the type of RINs (Renewable Energy Credits) they are eligible for. RNG produced from a dairy digester falls under the D3 RIN category (cellulosic biofuel). The Q4 2019 average D3 RIN price was \$0.88<sup>9</sup>. The RFS credit price for dairy digester biomethane can therefore be calculated by:

$$P_{RFS, \text{biomethane}} = \frac{\$0.88}{RIN} \times \left( \frac{1}{77,000} \right) \frac{RIN}{Btu} \times 10^6 \frac{Btu}{MMBtu} = \frac{\$11.4}{MMBtu}$$

E-methane produced from the P2G process doesn't not fall under any of the pre-approved pathways and as such is not eligible for RFS credits. Assuming biocrude oil falls under the D4 RIN category (biomass-based diesel), the RFS credit for biocrude oil can be estimated by:

$$P_{RFS, \text{biocrude oil}} = \frac{\$0.50}{RIN} \times \left( \frac{1}{77,000} \right) \frac{RIN}{Btu} \times 10^6 \frac{Btu}{MMBtu} = \frac{\$6.5}{MMBtu}$$

where \$0.50<sup>9</sup> is the Q4 2019 D4 RIN price.



## LCFS

Credits generated under the LCFS are based on the carbon intensity (CI, in  $\text{gCO}_2/\text{MJ}$ ) of the fuel production pathway. Credits are generated when the CI of the renewable fuel pathway is lower than that of the conventional fuel it replaces. The LCFS was developed to reduce the carbon intensity of California's transportation sector. In this study, the methane produced is compressed and sold as CNG to be used in CNG vehicles, as a direct replacement to gasoline. Every year, CARB sets carbon intensity benchmarks for gasoline and diesel fuel. The benchmark CI decreases linearly every year to meet the 20% average CI reduction (relative to 2010) by 2030 target<sup>10</sup>. The 2019 compliance year carbon intensity benchmark of gasoline, based on the average crude oil delivered to California refineries and average California refinery efficiencies (CARBOB model), is  $93.23 \text{ g CO}_{2\text{e}}/\text{MJ}$ <sup>10</sup>. Bio crude oil is assumed to be sold as a diesel substitute, and its CI will be compared to the CI benchmark (2019 compliance year) for California ULSD diesel,  $94.17 \text{ g CO}_2/\text{MJ}$ <sup>10</sup>.

For the dairy digester, we assumed a CI of  **$-276 \text{ gCO}_{2\text{e}}/\text{MJ}$**  of biomethane produced. This value accounts for the avoided emissions associated with methane offsets by treating manure and was calculated using the CA-GREET 1.8b model<sup>11,12</sup> for the CalBio LLC dairy digester in Bakersfield, California. Note that a full life cycle analysis is required to determine an accurate CI of the 26 individual digesters in this study, however, this was considered beyond the scope of this study. As a starting point, the carbon intensity of CalBio LLC was assumed instead, which falls between the typical range of dairy digesters systems carbon intensities in the United States<sup>13</sup>.

The CI of the electricity used for the power-to-gas process depends on the source of electricity used in electrolysis. A renewable source such as wind or solar results in a zero-carbon intensity. The CI of the NY electric grid, based on the current fuel mix is estimated by:

$CI_{NY \text{ electric grid}}$

$$= \frac{\text{net } CO_2 \text{ emissions from electric power generation (2018)}}{\text{net electricity generation (2018)}} = \frac{2}{13}$$

$$= 58.6 \text{ gCO}_2/\text{MJ}$$

Data for electricity generation and related emissions were taken from the EIA<sup>14</sup>. The CO<sub>2</sub> sourced from AD and HTL for the biomethanation process is assumed to have no carbon burden since the carbon is sourced from biogenic sources.

The allocation of carbon benefits (avoidance) and burdens (emission) among the different renewable fuels produced depend on the system boundaries assumed. In this study, three cases were evaluated: (A) AD, (B) AD-P2G and (C) AD-P2G-HTL. For simplicity, the allocation of carbon benefits/burdens to the produced transportation fuels (biomethane, e-methane and bio oil) was done using a lumped-process approach, where AD biogas upgrading, P2G and HTL are lumped within a single process with biogas and electric power as inputs (Table S2).

Table S2 – System inputs and transportation fuels outputs for the three cases evaluated. Calculations were performed per unit biogas input (1 Nm<sup>3</sup>).

	Inputs		Outputs		
	Biogas (Nm <sup>3</sup> )	Electric Power (MJ)	Bio-methane (Nm <sup>3</sup> )	E-methane (Nm <sup>3</sup> )	Biocrude oil (Nm <sup>3</sup> )
Case A	1	N/A	0.56 (20.8 MJ)	N/A	N/A
Case B	1	32	0.56 (20.8 MJ)	0.44 (16.3 MJ)	N/A
Case C	1	44	0.56 (20.8 MJ)	0.61 (22.8 MJ)	3*10 <sup>-4</sup> (7.84 MJ)

The carbon benefit (avoided emissions) of producing 1 Nm<sup>3</sup> of biogas is given by:

$$\text{carbon benefit} = -276 \frac{\text{gCO}_2}{\text{MJ}} \times 20.8 \text{ MJ} = -5,730 \text{ g CO}_2$$

The carbon burden (emissions) of using NY grid electricity (58.6 g CO<sub>2</sub>/MJ) for the electrolysis process in cases B and C is shown below.

$$\text{carbon burden}_B = 58.6 \frac{\text{gCO}_2}{\text{MJ}} \times 32 \text{ MJ} = 1,856 \text{ g CO}_2$$

$$\text{carbon burden}_C = 58.6 \frac{\text{gCO}_2}{\text{MJ}} \times 44 \text{ MJ} = 2,587 \text{ g CO}_2$$

The carbon burden of using renewable electricity is 0 g CO<sub>2</sub>. Allocation was conducted on an energy-basis. For case A, all the carbon benefit from treating the manure (-5,730 g CO<sub>2</sub>) is allocated to biomethane. In case B, that carbon benefit is split among biomethane and e-methane based on the embedded energy in each product. In case C, the carbon benefit is split among biomethane, e-methane and bio crude oil, on an energy basis. The carbon burden of the electric power is similarly split among the produced fuels for cases A, B and C. Carbon intensity of the fuels is then obtained by dividing the carbon allocation values for each fuel by the energy embedded in that fuel. The results are tabulated in Tables S3 and S4.

Table S3 - Carbon allocation for the renewable fuels produced.

Carbon allocation (g CO <sub>2</sub> )	Bio-methane	E-methane	Biocrude oil
Case A	-5730	N/A	N/A
Case B			
NY-grid power	-2168 <sup>a</sup>	-1706	N/A
Renewable power	-3207 <sup>b</sup>	-2524	N/A
Case C			
NY grid power	-1270	-1393 <sup>c</sup>	-479
Renewable Power	-2316	-2540 <sup>d</sup>	-874

$$\begin{aligned} \text{a: } & (-5730 + 1856) \times \left( \frac{20.8}{20.8 + 16.3} \right) = -2168 \text{ g CO}_2 \\ \text{b: } & -5730 \times \left( \frac{20.8}{20.8 + 16.3} \right) = -3207 \text{ g CO}_2 \\ \text{c: } & (-5730 + 2587) \times \left( \frac{22.8}{20.8 + 22.8 + 7.84} \right) = -1393 \text{ g CO}_2 \\ \text{d: } & -5730 \times \left( \frac{22.8}{20.8 + 22.8 + 7.84} \right) = -2540 \text{ g CO}_2 \end{aligned}$$

Table S4 – Carbon intensity of the renewable fuels produced

Carbon Intensity (g CO <sub>2</sub> /MJ)	Bio-methane	E-methane	Biocrude oil
Case A	-276	N/A	N/A
Case B			
NY-grid power	-104	-104	N/A
Renewable power	-154	-154	N/A
Case C			
NY grid power	-61	-61	-61
Renewable Power	-112	-112	-112

For cases B and C, we also considered a subcase where the carbon burden of using grid electricity is allocated to e-methane only. The resulting CI values are reported in Table S5. In this subcase, the CI of e-methane increases drastically, while that of biomethane and biocrude oil would equal that of using renewable power.

Table S5 – Carbon allocation and carbon intensities of the produced fuels for cases B and C, in the subcase where the carbon burden from using grid electricity is only allocated to e-methane.

	Bio-methane	E-methane	Biocrude oil
Carbon allocation (g CO <sub>2</sub> )			
Case B	-3207	-667 <sup>a</sup>	N/A
Case C	-2316	47 <sup>b</sup>	-874
Carbon intensity (g CO <sub>2</sub> /MJ)			

Case B	-154	-41	N/A
Case C	-112	2	-112
a:	$-5730 \times \left( \frac{16.3}{20.8 + 16.3} \right) + 1856 = -667$		
b:	$-5730 \times \left( \frac{22.8}{20.8 + 22.8 + 7.84} \right) + 2587 = 47$		

The CI of shipping the produced RNG and biocrude oil to California, by pipeline and rail respectively, is added to the CI values in Tables S4 and S5 to get the overall CI of the delivered fuels. The LCFS credits (\$/MJ) for RNG and biocrude oil can be then calculated by:

$$P_{LCFS, RNG} = \left( 93.23 \frac{gCO_2}{MJ} - (CI_{RNG} + CI_{pipeline}) \right) \times 10^{-6} \frac{MT}{g} \times \frac{\$196}{MT CO_2}$$

$$P_{LCFS, biocrude\ oil} = \left( 94.17 \frac{gCO_2}{MJ} - (CI_{biocrude\ oil} + CI_{rail}) \right) \times 10^{-6} \frac{MT}{g} \times \frac{\$196}{MT CO_2}$$

where \$196/MT CO<sub>2</sub> is the average LCFS credit price for Q4 2019. The LCFS credit prices (\$/MT CO<sub>2</sub>) can be found online from the monthly LCFS credit transfer activity reports<sup>15</sup>. 93.23 g CO<sub>2</sub>/MJ and 94.17 gCO<sub>2</sub>/MJ are the carbon intensity benchmarks of California gasoline (CARBOB) and diesel (ULSD), respectively.  $CI_{RNG}$  and  $CI_{biocrude\ oil}$  are the carbon intensities of RNG (biomethane or e-methane) and biocrude oil produced, respectively.  $CI_{pipeline}$  and  $CI_{rail}$  were calculated using emission factors obtained from the tier 1 CA GREET model for AD biomethane<sup>16</sup> (4.08 g CO<sub>2</sub>/MMBtu/mile) and biodiesel<sup>16</sup> (0.1 g CO<sub>2</sub>/gal/mile). A transportation distance of 2,800 miles between New York and California was used for calculations.

Since both the LCFS and RFS credit programs are stackable<sup>17-19</sup>, the premiums for RNG and biocrude oil (selling price) can be calculated by:

$$P_{biomethane} = P_{LCFS} + P_{RFS} + P_{HH}$$

$$P_{emethane} = P_{LCFS} + P_{HH}$$

$$P_{biocrude\ oil} = P_{LCFS} + P_{RFS} + P_{ULSD}$$

where  $P_{HH}$  and  $P_{ULSD}$  are the natural gas (NG) and Ultra-Low-Sulfur Diesel (ULSD) fuel spot prices on the US commercial energy market (\$/MJ). The Henry Hub NG spot price of \$2.4/MMBtu was used for  $P_{HH}$  as reported by the New York Mercantile Exchange (NYMEX) for Q4 2019<sup>20</sup>. The Q4 2019 New York Harbor ULSD spot price<sup>21</sup> of \$1.95/gal (\$15.3/MMBtu) was used for  $P_{ULSD}$ .

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