

## 26th CIRP Life Cycle Engineering (LCE) Conference

## Greenhouse Gas Emissions of Transportation Fuels from Shale Gas-Derived Natural Gas Liquids

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Production of natural gas liquids (NGLs) in the United States is expanding rapidly and production now exceeds domestic demand. An emerging use of NGLs is the production of transportation fuels, however, the greenhouse gas (GHG) emissions of these fuels may limit their access to some markets affected by policies that consider these emissions. This work estimates well-to-tank GHG emissions of NGL-based transportation fuels and compares these estimates to GHG emissions from conventional gasoline production. The emission estimates, and their magnitude relative to well-to-tank GHG emissions for conventional petroleum fuels, are highly sensitive to NGL fuel production scenarios, co-product treatment methods, and feedstock source.

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**Keywords:** Shale gas; natural gas liquids; transportation fuels; greenhouse gas emissions; life-cycle assessment; well-to-tank**1. Introduction**

Recently, the nature of hydrocarbon production in the United States has changed, making possible new routes to the production of transportation fuels. In the United States, production of natural gas and natural gas plant liquids (NGPLs or NGLs) using horizontal drilling and hydraulic fracturing has expanded rapidly. Overall production of natural gas [1] increased from 24.6 trillion cubic feet (tcf) in 2007 to 33.2 tcf in 2017 and production of NGLs (primarily ethane, propane, and butane) [2] increased from 600 million barrels (bbl) per year in the period from 1980–2007 to 1277 million bbl per year in 2016. Ethane and propane are now being produced at rates that exceed United States domestic demand for them; they are beginning to be exported.

An alternative to the export of these NGLs could be their transformation into liquid transportation fuels. Ridha et al. [3] has recently described a conceptual process for transformation

of NGLs into liquid transportation fuels, shown in Figure 1. In this process, light alkanes are dehydrogenated, then oligomerized to yield gasoline and other fuel products. However, the sale of such unconventional transportation fuels could be limited in some market segments by their life-cycle greenhouse gas (GHG) emissions. For example, Section 526 of the Energy Independence and Security Act of 2007 (EISA) [4] requires that alternative transportation fuels sold to the federal government must have lower or equal life-cycle GHG emissions compared to baseline petroleum-based fuels, the value of which was set by a National Energy Technology Laboratory (NETL) analysis based on gasoline sold or distributed in the United States as transportation fuels in 2005 [5]. Additionally, the California Low Carbon Fuel Standard [6] limits the sale of transportation fuels in California based on life-cycle GHG emissions. Therefore, it is important to understand the life-cycle GHG emissions of fuel production pathways that may emerge from expanded NGL production.

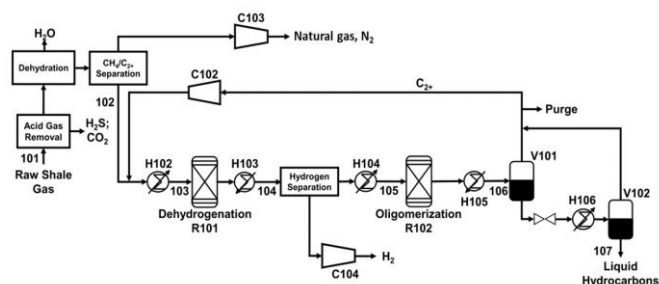


Fig. 1. A conceptual transformation of NGLs into transportation fuels (Process I) [3].

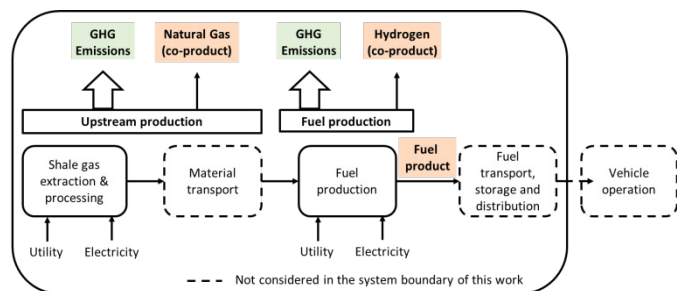


Fig. 2. Life-cycle stages and system boundary of the study.

This work will examine the life-cycle GHG emissions of the hydrocarbon fuels produced from NGLs and compare these GHG burdens to baseline petroleum-based fuels. GHG emissions in oil and gas production operations (upstream production) are combined with the emissions associated with the liquid fuel manufacturing process (fuel production) to arrive at the total well-to-tank (WTT) emission estimates, as shown in Figure 2. Because GHG emissions can vary significantly among production regions, the analyses will initially be performed by assuming a national average value of upstream GHG burdens. The estimated emissions will then be compared to conventional transportation fuels manufactured from petroleum-based feedstocks. The sensitivity of the GHG burdens to variability in emissions associated with different gas production regions, different assumptions concerning the fuel manufacturing processes, and different assumptions in the life-cycle calculations will then be examined.

## 2. Methodology

### 2.1. Life-cycle stages and system boundary

Figure 2 shows the life-cycle stages and system boundary investigated in this study. The same five life-cycle stages used by the National Energy Technology Laboratory (NETL) [5] in their evaluation of baseline, petroleum-derived fuels are used in this work. A WTT analysis, omitting the vehicle operation stage is considered in this analysis. Emissions from the operation stage are not included in comparisons, because the hydrocarbon fuels produced using NGL feedstocks are assumed to have a composition similar to conventional gasoline, and thus generate GHG emissions similar to conventional gasoline when combusted. Therefore, even though 80% of life-cycle GHG emissions of petroleum-based gasoline are generated in the vehicle operation stage [5], and

the same might be assumed for NGL-based fuels, the vehicle operation stage is not considered in the system boundary, in order to highlight the differences between the fuel systems. Transportation of both raw materials and fuel products are not considered, primarily because GHG emissions from raw materials / fuel product transportation are relatively low compared to the emissions from the other life-cycle stages, as evaluated by NETL [5]. In addition, differences between the transportation of NGL-based fuels and petroleum-based fuels may be small since, under some scenarios, NGL production and processing may occur in locations similar to the locations of current petroleum production and refining operations.

In this study, the WTT GHG emissions are a combination of upstream and fuel production components, as shown in Figure 2. The upstream production separates NGLs, used as feedstock for transportation fuel production, from pipeline quality natural gas, which is treated as a co-product. Figure 1 shows the fuel production process (from NGLs to fuel), in which NGLs from upstream production will undergo catalytic dehydrogenation and oligomerization to produce hydrocarbon fuels as the main product and hydrogen as a co-product. All operations associated with upstream shale gas production and processing emit methane (a potent GHG). Fuel combustion emissions (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emissions from process energy consumption) from shale gas production and processing and non-combustion emissions (CO<sub>2</sub> emissions from equipment venting) add additional burdens. For the fuel production component, process energy demand is the only contributor to GHG emissions. The GHG emissions (CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O) contributed by process energy demand in fuel production are assumed to come from the combustion of natural gas (source of heating utility) and generation of electricity required by the process.

### 2.2. GHG impact assessment

All the GHG emissions in this study are weighted based on their global warming potentials over a 100-year period, reported in the units of grams of equivalent emissions of carbon dioxide (g CO<sub>2</sub>e). The global warming potential factors for methane and N<sub>2</sub>O applied in this study are the international standard values published in The Intergovernmental Panel on Climate Change (IPCC) in 2007 based on the 100-year time frame [7]. Recently the IPCC has released higher, updated, global warming potentials, however, in order to be consistent with the impact assessment method developed by NETL [5] for benchmark fuels, the global warming potentials for methane of 25 and N<sub>2</sub>O of 298 are assumed in this work. The life-cycle GHG emissions are normalized by the lower heating value (LHV) of the heat of combustion of the liquid hydrocarbon fuels and are reported as grams of CO<sub>2</sub>e emissions per megajoule of LHV of hydrocarbon fuels (g CO<sub>2</sub>e / MJ hydrocarbon fuels).

### 2.3. Co-product treatment methods

Allocation methods can be used in LCA when multiple products are produced to divide energy and environmental burdens among co-products. Allocation is commonly based

on energy, mass, or market value, depending on the properties of the products. An energy-based allocation is applied in this analysis since all the products are expected to be used based on their energy content. GHG emissions from upstream production are allocated among natural gas (the co-product from upstream production), hydrogen (the co-product from NGL dehydrogenation), and liquid hydrocarbon fuels (the main product). GHG emissions from the fuel production are allocated between liquid hydrocarbon fuels and hydrogen. Other co-product treatment methods could be assumed (e.g., displacement), however, use of hydrogen for purposes other than as a fuel would require co-location of hydrogen production and use or reliance on a hydrogen transport infrastructure that is currently limited. The GHG emissions allocated to hydrocarbon fuels are calculated with the following equation:

*WTT emissions allocated to hydrocarbon fuels*

$$= \left( \frac{\text{Upstream}}{\text{emissions}} \right) \times \frac{\text{LHV of fuels}}{\text{LHV of NG} + \text{LHV of H}_2 + \text{LHV of fuels}} + \left( \frac{\text{Fuel production}}{\text{emissions}} \right) \times \frac{\text{LHV of fuels}}{\text{LHV of H}_2 + \text{LHV of fuels}}$$

where NG refers to natural gas; fuels specifically refer to the hydrocarbon fuel product.

### 3. Input data and key parameters

#### 3.1. Upstream production

Methane emissions from all operations associated with shale gas recovery and processing are estimated by using the national average of upstream methane emission rates. Alvarez et al. [8] investigated the total methane emissions in 2015 from the U.S. oil / natural gas supply chain and reported a production-normalized emission rate of 2.3% (+0.4/-0.3%) by normalizing the total methane emissions from upstream activities by annual gross natural gas production. Littlefield et al. [9] reported 1.7% (+0.5/-0.4%) of the methane in natural gas was emitted between extraction and delivery based on a series of methane emission measurements focusing on the U.S. natural gas supply chain. In this study, we assume that 2% of the methane in raw shale gas is emitted to the atmosphere, recognizing that, as reported by Alvarez et al., [8] there is significant variability in emissions between different production regions. Methane emissions are then converted to CO<sub>2</sub>e emissions to arrive at the GHG impact estimates. The GHG burdens due to process energy demand for shale gas recovery and processing also contribute to upstream emissions and are calculated based on emission factors provided in The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation Model (GREET®) [10]. Feedstock and product profiles (synthesized and provided by Ridha et al. [3]) and emission factors involved in upstream calculations are shown in Table 1.

Table 1. Feedstock and product profiles and emission factors for a shale gas processing facility handling 97.06 MMSCFD of raw shale gas.

Feedstock	
Raw shale gas	97.06 MMSCFD [3]
Products	
Natural gas	56.23 MMSCFD [3]
NGLs	34.81 MMSCFD [3]
Emission factors	
Methane emission rate	2% of methane in raw shale gas
Shale gas recovery	2.13 g CO <sub>2</sub> e / MJ natural gas [10]
Shale gas processing	2.47 g CO <sub>2</sub> e / MJ natural gas [10]

#### 3.2. Fuel production

GHG emissions from fuel conversion processes estimated in this study are exclusively contributed by process energy demand. Ridha et al. [3] developed process models for NGL conversion to liquid hydrocarbon fuels and hydrogen. Materials and energy requirements and product yields from these simulations are shown in Table 2. These material and energy flows are the key inputs for GHG emissions calculations. The hydrocarbon fuel product considered in this study is assumed to be in the gasoline range (C4-C12). The other key parameters involved in GHG emissions estimation are summarized in Table 3.

Table 2. Energy requirements and product profiles for a NGLs-to-fuel facility processing 34.81 MMSCFD of NGLs [3].

Feedstock	
NGLs	34.81 MMSCFD
Energy requirements	
Electricity	0.62 MW / MMSCFD of NGLs
Heating utility	1.84 MW / MMSCFD of NGLs
Products	
Hydrocarbons	17.98 kmol / hr · MMSCFD of NGLs
Hydrogen	36.95 kmol / hr · MMSCFD of NGLs

Table 3. Key parameters in GHG emissions estimation.

Hydrocarbon stream profile	
fuel gas (C1-C3) <sup>a</sup>	3.38% (mole fraction) [3]
hydrocarbon fuels (C4-C12)	96.62% (mole fraction) [3]
Molar mass of hydrocarbon fuels	83.02 g/mol [3]
Lower heating values (LHVs)	
Natural gas	1.04 MJ / SCF [10]
NGLs <sup>b</sup>	2.11 MJ / SCF [11]
Hydrogen	0.31 MJ / SCF [10]
Hydrocarbon fuels <sup>c</sup>	43.24 MJ / kg [10]
Density	
Hydrocarbon fuels	2.819 kg / gal [10]
Hydrogen	0.003 kg / ft <sup>3</sup> (at 32 °F, 1atm) [10]
Natural gas	0.022 kg / ft <sup>3</sup> (at 32 °F, 1atm) [10]
Global warming potential over 100 years	
CO <sub>2</sub>	1 [7,17]
CH <sub>4</sub>	25 [7,17]

N <sub>2</sub> O	298 [7,17]
Emission factors for process energy	
Electricity	149.73 g CO <sub>2</sub> e / MJ of electricity required [10]
Heating from combustion of natural gas	70.66 g CO <sub>2</sub> e / MJ of heat required (assuming energy efficiency of steam boilers as 80%) [10]

Notes: <sup>a</sup> Fuel gas is primarily dissolved in the hydrocarbon stream but will eventually be either emitted, flared or recovered for its fuel value. <sup>b</sup> LHV of the NGLs is calculated by dividing the total energy flow rate (sum of energy flow rates of each component) by the total volumetric flow rate of the NGL stream; the energy flow rate of each component is derived by multiplying the mass based LHV of each component (LHV of each component is calculated by subtracting the heat of vaporization of water from the higher heating value of each component reported by National Institute of Standards and Technology [11]) by the mass flow rate of each component; the mass flow rate of each component is calculated based on the total volumetric flow rate of NGL stream, mole fraction of each component, and density of each component; the density of each component is estimated by ideal gas law. <sup>c</sup> The number reported here is the LHV of gasoline blendstock reported in the GREET model; LHV calculated for hydrocarbon fuels is 44.30 MJ/kg (same calculation steps as for NGLs). The hydrocarbon fuel product is assumed to be gasoline blendstock, therefore the LHV and density of gasoline blendstock are applied for the hydrocarbon fuels in calculations.

## 4. Results and discussion

### 4.1. GHG emissions

For the production of 1 MJ hydrocarbon fuel product, the feedstock requirements, process energy demand, and production rates of all the products are estimated, and are then combined with emission factors to arrive at the GHG emissions estimate. The emission allocation factors among the products are calculated based on the energy content of the products as shown in Table 4. 44% of GHG emissions from upstream production, and 88% of the emissions from fuel production are allocated to hydrocarbon fuels. The GHG emissions are reported by emission category in Table 5. GHG emissions from upstream and fuel production components assigned to the hydrocarbon fuels are 9.6 and 11 g CO<sub>2</sub>e / MJ hydrocarbon fuels respectively, with a total WTT emissions of 21 g CO<sub>2</sub>e / MJ hydrocarbon fuels.

In order to benchmark the GHG emissions of transportation fuels derived from NGLs, the results are compared to the average WTT GHG emissions from conventional petroleum-based gasoline sold or distributed in the United States as transportation fuels in 2005, developed by NETL, known as the NETL 2005 Petroleum Baseline [5].

Table 4. Energy content (LHV) and energy-based allocation factors of upstream and fuel production segments for producing 1 MJ hydrocarbon fuel product.

Product and co-products	Energy content (LHV) per 1 MJ hydrocarbon fuels	Upstream production	Fuel production
Natural gas (co-product)	1.1 MJ	0.49	N/A
Hydrogen (co-product)	0.14 MJ	0.06	0.12
Hydrocarbon fuels	1 MJ	0.44	0.88

Table 5. GHG emissions of each emission category for production of 1 MJ hydrocarbon fuels and GHG emissions allocated to hydrocarbon fuels.

Emission category	Total emissions (g CO <sub>2</sub> e)	Allocated emissions (g CO <sub>2</sub> e / MJ hydrocarbon fuels)
Upstream production		
Methane emissions	10	4.5
Shale gas production	5.3	2.4
Shale gas processing	6.1	2.7
Upstream total	22	9.6
Fuel conversion		
Electricity	5.3	4.7
Process heating	7.5	6.6
Fuel conversion total	13	11
Total emissions		21

Figure 3 shows the comparison of GHG emissions between NGL-based fuel production estimated in this study and the NETL 2005 Petroleum Baseline. For upstream production, GHG emissions from NGL fuel production are 40% higher than from conventional gasoline production, mainly because of the high methane emissions associated with upstream shale oil/gas operations, which are double the estimates for petroleum production as documented in the NETL assessment [5]. GHG emissions from NGL fuel production are 11 g CO<sub>2</sub>e / MJ fuel, 18% higher than from conventional gasoline production (9.3 g CO<sub>2</sub>e / MJ fuel). The estimate for the NGL fuel production may be biased high relative to the conventional gasoline production, since the conventional production of gasoline is highly optimized, efficient and integrated, but the emerging NGLs-to-fuel pathway is still being developed. Total WTT GHG emissions from NGL fuel pathway are 31% higher than the NETL 2005 Petroleum Baseline (16 g CO<sub>2</sub>e / MJ fuel). However, the conventional baseline value may change with time due to changes in the U.S. petroleum sector. Cooney et al. [12] reported a 4.1% increase between NETL 2005 Petroleum Baseline and the baseline results for 2014. Also, WTT GHG emissions from petroleum-based gasoline production are reported as 19 g CO<sub>2</sub>e / MJ fuels in GREET1\_2017 [10]. Therefore, the emerging NGLs-to-fuel pathway could be comparable to conventional gasoline production in terms of GHG emissions, with better integrated and optimized processes.

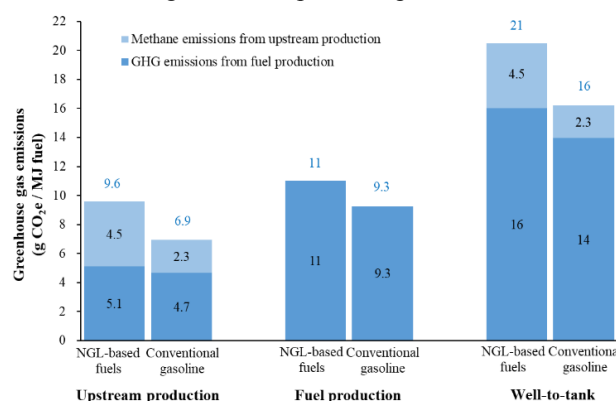


Fig. 3. GHG emissions from NGL-based fuel production compared to NETL 2005 Petroleum Baseline [5].

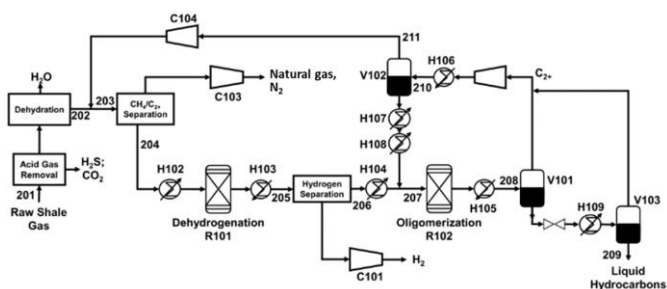


Fig. 4. A second conceptual transformation of NGLs into transportation fuels (Process II) [3].

#### 4.2. Sensitivity analysis

Estimated GHG burdens are sensitive to assumptions made for both upstream production and downstream fuel production. To assess sensitivity of GHG burdens to the NGL fuel production configuration used in this work, a second fuel production configuration was evaluated. In addition to the fuel production process used as the baseline in this work (Process I), Ridha et al. [3] also proposed another process (Process II), with a different natural gas / NGLs separation technology and with different recycle loop configurations, shown in Figure 4. The estimation of GHG emissions from Process II follows the same steps as Process I, assuming the same raw shale gas feed rate (97.06 MMSCFD), as in Process I. The comparison of GHG emissions for producing 1 MJ hydrocarbon fuels for the 2 processes is shown in Table 6.

GHG emissions from upstream NGL production of both processes are assumed to be identical. Electricity requirements and process heating duties contribute to GHG emissions from the fuel conversion process. Heating duties of the two processes are assumed to be made similar through process heat integration, approximately equal to the heat of reaction for dehydrogenation, thus contributing to similar GHG emissions (7.5 and 7.4 g CO<sub>2</sub>e for producing 1 MJ hydrocarbon fuels in Processes I and II, respectively). For the production of 1 MJ hydrocarbon fuels from NGLs, Process I requires more electricity, and therefore generates more GHG emissions (5.3 g CO<sub>2</sub>e) compared to Process II (4.6 g CO<sub>2</sub>e). Differing electricity requirements therefore are a key factor in the variation in GHG emissions between the two processes. A second factor is the rate of production of the co-product hydrogen. 93% of GHG emissions from the fuel production segment are allocated to hydrocarbon fuels in Process II, compared to 88% in Process I, because Process I has higher hydrogen production and recovery rates [3]. The overall WTT GHG emissions of Process II are 20 g CO<sub>2</sub>e / MJ of hydrocarbon fuels, similar to the baseline process.

In addition to different fuel production configurations, the use of co-product treatment methods will impact estimated GHG burdens. In this study, the energy allocation method is used and GHG emissions are allocated among co-products based on their energy values as discussed in the methodology section, however, other allocation bases could be applied. Table 6 lists the energy, mass and market values of the products manufactured per 1 MJ production of hydrocarbon fuels. Table 7 shows the energy, mass and market value allocation factors for upstream production and fuel production

segments. Mass or market value rather than energy-based allocations would increase the GHG burdens of the NGL-based fuels.

In addition to uncertainties in the GHG burdens associated with fuel processing configurations and allocation methods, there is uncertainty in the estimation of upstream methane emissions. Production-normalized methane emission rates vary significantly among oil and gas production regions due to reservoir structures, reservoir fluids types, gas extraction technologies, and other factors. Among the largest shale basins in the U.S., the production-normalized methane emission rate in the Barnett Shale is 1.5% [16], which is over 10 times greater than the emission rate in the Marcellus Shale (0.13%) [17]. GHG emissions could be reduced to 17 g CO<sub>2</sub>e / MJ hydrocarbon fuels if NGLs come from shale gas recovered and converted to transportation fuels in the Marcellus Shale. Therefore, feedstock origin will impact the GHG emissions of NGLs based transportation fuels and might lead to >±20% changes in estimated GHG burdens.

Table 6. Comparison of GHG emissions for producing 1 MJ hydrocarbon fuels between two NGL conversion process configurations.

GHG emissions	Process I	Process II
Upstream production		
Methane emissions (g CO <sub>2</sub> e)	10	10
Shale gas recovery (g CO <sub>2</sub> e)	5.3	5.2
Shale gas processing (g CO <sub>2</sub> e)	6.1	6.0
Upstream total (g CO <sub>2</sub> e)	22	21
Fuel conversion		
Electricity (g CO <sub>2</sub> e)	5.3	4.6
Process heating (g CO <sub>2</sub> e)	7.5	7.4
Fuel conversion total (g CO <sub>2</sub> e)	13	12
GHG emissions allocated to hydrocarbon fuels		
Upstream (g CO <sub>2</sub> e / MJ fuels)	9.6	8.7
Fuel production (g CO <sub>2</sub> e / MJ fuels)	11	11
WTT total (g CO <sub>2</sub> e / MJ fuels)	21	20

Table 7. Energy, mass and market value production of co-products per production of 1 MJ hydrocarbon fuels.

Products	Energy (MJ)	Mass (g)	Market value (× 10 <sup>-3</sup> USD, based on average price in 2017)
Natural gas	1.1	22	4.5 [13]
Hydrogen	0.14	1.2	3.5-11 <sup>a</sup> [14]
Hydrocarbon fuels	1	23	20 [15]

<sup>a</sup> depending on target markets of hydrogen

Table 8. Allocation factors of upstream production and fuel production segments based on energy, mass, and market values of co-products.

Products	Energy allocation	Mass allocation	Market value allocation <sup>a</sup>
Upstream production			
Natural gas	0.49	0.48	0.13-0.16
Hydrogen	0.06	0.03	0.13-0.31
Hydrocarbon fuels	0.44	0.49	0.56-0.71
Fuel production			

Hydrogen	0.12	0.05	0.15-0.35
Hydrocarbon fuels	0.88	0.95	0.85-0.65

<sup>a</sup> depending on target markets of hydrogen

### 4.3. Emission reduction possibilities for fuel production

Combustion of natural gas for process heating and electricity generation for compressors in the recycle stream and hydrogen stream are the main contributors to GHG emissions in the fuel production segment. Heating duties in fuel production segment could be minimized to the heat of reaction for dehydrogenation through process heat integration. In addition, complete recovery of the C1-C3 product stream could further reduce process energy demand. Process electricity requirements could be reduced by decreasing compressor loads in the recycle stream. This could be achieved by increasing the per pass fuel conversion rate of the oligomerization reaction and by optimizing product flash conditions. In addition, increasing the hydrogen recovery rate by improving the selectivity of hydrogen membrane separation technology can also help to reduce GHG emissions allocated to the hydrocarbon fuel product.

GHG emissions from the upstream production segment depend significantly on feedstock sources and can be reduced if feedstocks from regions with low GHG emissions are used.

## 5. Conclusion

GHG emissions of transportation fuels produced from NGLs derived from shale gas were estimated on a WTT basis accounting for upstream and fuel production segments. WTT GHG emissions of the base case process are 21 g CO<sub>2</sub>e / MJ fuel, 31% higher than the NETL 2005 petroleum baseline for the production of conventional gasoline from refined petroleum. WTT GHG emissions of hydrocarbon fuels from NGLs, however, are highly sensitive to NGL fuel production scenarios, co-product treatment methods, and also feedstock source because of the variabilities in upstream emissions among production regions due to reservoir structures, reservoir fluids types, gas extraction technologies, and other factors. These factors can render NGL-derived fuel WTT GHG emissions higher or lower than conventional petroleum fuels. Furthermore, advancements and optimization of the NGL conversion technology can drive down WTT GHG emissions of NGL-derived fuels.

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