

Well-to-Wheels Emissions of Greenhouse Gases and Air Pollutants of Dimethyl Ether from Natural Gas and Renewable Feedstocks in Comparison with Petroleum Gasoline and Diesel in the United States and Europe

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ABSTRACT

Dimethyl ether (DME) is an alternative to diesel fuel for use in compression-ignition engines with modified fuel systems and offers potential advantages of efficiency improvements and emission reductions. DME can be produced from natural gas (NG) or from renewable feedstocks such as landfill gas (LFG) or renewable natural gas from manure waste streams (MANR) or any other biomass. This study investigates the well-to-wheels (WTW) energy use and emissions of five DME production pathways as compared with those of petroleum gasoline and diesel using the Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET®) model developed at Argonne National Laboratory (ANL). The five DME pathways include 1) fossil NG with large-scale DME plants, 2) methanol from fossil NG with large-scale plants for both methanol and DME (separately), 3) LFG with small-scale DME plants, 4) manure-based biogas with small-scale DME plants, and 5) methanol from black liquor gasification with small-scale DME plants. This study analyzes DME production and use in the U.S. and Europe, and in two vehicle classes (light and heavy duty vehicles [LDVs and HDVs]). The WTW results show significant reductions in fossil fuel consumption and greenhouse gas (GHG) emissions by DME compared to gasoline and diesel if DME is produced from LFG and manure-based biogas. When methanol from black liquor is used for DME production, there are reductions in GHG emissions, though smaller than DME produced from LFG and MANR. Meanwhile, fossil NG-based DME produced in large-scale DME plants or from NG-based methanol shows GHG emissions at the similar level as petroleum diesel does.

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INTRODUCTION

Dimethyl ether (DME), the simplest form of ether, is an alternative to conventional diesel fuel for use in compression-ignition engines. A

key advantage of using DME as an alternative to diesel is significantly low particulate emissions due to its lack of carbon-to-carbon bonds, which could reduce or eliminate the need for expensive

diesel particulate filters or after-treatment systems. DME also has a high cetane number while the energy efficiency and power ratings of DME and diesel engines are virtually the same [1]. Also, DME is compatible with current diesel engines with minor modifications, and could share modified liquefied petroleum gas (LPG) infrastructures for storage, transportation, and distribution, which are already widespread [2]. In 2014, the global DME market size is estimated at 34 million barrels per year. Note that a large portion of current DME market is for non-transportation uses. If all DME produced currently were used as transportation fuels, it could substitute about 2.5% of diesel consumption (1.36 billion barrels per year) [3]. In Europe, North America, and Asia, a number of DME vehicle fleets have been demonstrated [4, 5, 6, 7, 8, 9, 10, 11, 12].

DME is produced by dehydration of methanol, which is now largely produced from natural gas (NG). As demonstrated before [13], NG-derived DME does not reduce life-cycle fossil fuel use and greenhouse gas (GHG) emissions compared to petroleum gasoline and diesel. To reduce the life-cycle fossil fuel use and GHG emissions by DME and its application, alternative, renewable feedstocks for DME production are being considered, including biomass and biogas. Annual U.S. biogas resource is estimated at 370 PJ in the Biogas Opportunities Roadmap report [14]. Also, based on the Inventory of U.S. Greenhouse Gas Emissions and Sink by US Environmental Protection Agency (EPA), Argonne National Laboratory (ANL) estimates U.S. biogas resource to be 211 PJ per year excluding the landfill gas (LFG) currently used for energy recovery [15]. If used solely for DME production, the biogas resource estimates by Biogas Opportunities Roadmap and ANL could replace 6.2 GL and 3.5 GL of diesel per year or 3% and 2% of total diesel consumption in 2015, respectively. On the other hand, the U.S. Billion-Ton Update [16] estimate the 243 - 767 million dry tons of biomass will be available in 2030. If all of the biomass resource is used for DME, the total diesel replacement could be 24 - 75 GL per year, or 10 - 33% of total diesel consumptions in 2030.

Several studies have conducted life-cycle analysis (LCA) to evaluate the life-cycle impacts of DME from biomass and biogas feedstocks. For biomass gasification routes, Silalertruksa et al. [17] studied DME from rice straw in Thailand, and showed that life-cycle GHG emissions of the DME pathway were 25 gCO₂e/MJ. Similarly, Higo and Dowaki [18] performed a case study of DME production pathways from various woody and herbaceous biomass types in Japan and Papua New Guinea and estimated that the GHG intensities of these pathways ranged from 12 to 47 gCO₂e/MJ. Another study on DME LCA using Japanese cedar suggested 46 - 59 gCO₂e/MJ of DME [19]. Moreover, a recently published well-to-tank (WTT) report by JEC (a joint effort by European Commission's Joint Research Centre, EUCAR, and CONCAWE) included LCA of DME production from fossil NG and woody biomass [20]. The results showed that GHG emission intensities were ranged 89 - 98 gCO₂e/MJ for the NG-to-DME pathways depending on the plant operation conditions and the relative distances between the NG fields and the end use. The GHG emissions of biomass-based-DME (bio-DME) were as low as 6.6 gCO₂e/MJ. The GHG emissions for bio-DME in the JEC study were much lower than the estimates by other studies because the JEC study assumed that most process energy for biomass gasification and DME production was provided by biomass combustion while the other studies assumed by external fuels (electricity, NG, and oil). On the other hand, for DME from biogas, an LCA study by California Air Resources Board (CARB) estimated that GHG emissions of the biogas-based DME pathway were -5 gCO₂e/MJ as compared to 91 gCO₂e/MJ for DME from NG [2]. Also, Moghaddam et al. [21] estimated 11 gCO₂e/MJ for a similar DME pathway from biogas. In both studies, the system boundary starts from collected biogas, but does not include the biogas generation from organic waste. For black liquor-based DME, Joelsson and Gustavsson [22] estimated the GHG emissions changes in kg CO₂e of a chemical pulp mill by installing biorefinery. Landälv et al. [12] demonstrated that more than 500 tons of bio-DME from black liquor gasification has been produced and used for more than 1 million km in commercial service.

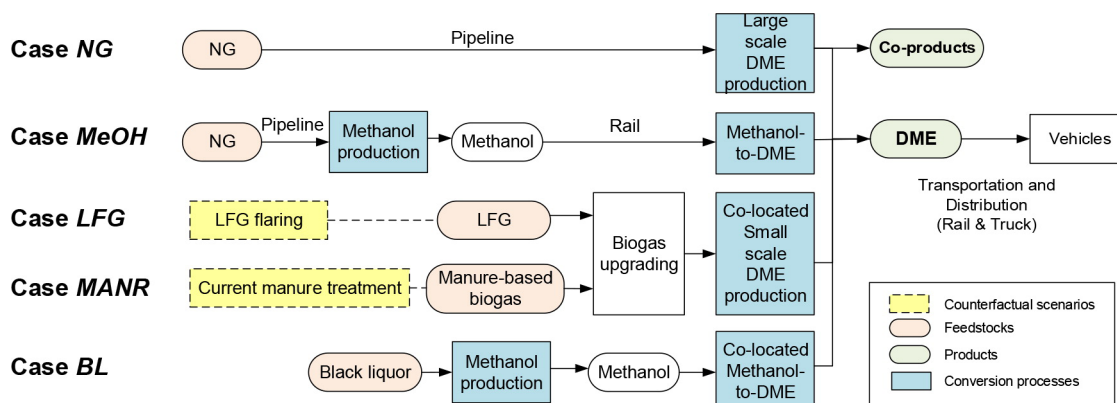


Figure 1. System boundary and key parameters of DME pathways

The fore-mentioned studies for DME from biogas and black liquor have limited scope (i.e., not including biogas generation from organic waste) or have inconsistent functional unit (i.e., per plant). To consistently and systematically evaluate DME GHG impacts, this study presents a comprehensive LCA on DME fuel from five production pathways using fossil and renewable feedstocks, and compares them with gasoline and diesel fuels. The feedstocks in this study include fossil NG, LFG, manure-based biogas and black liquor. For the fossil NG pathways, both integrated DME production from NG and separate DME production from methanol are considered. In order to reflect regional differences, the analysis is performed for two major markets - the US and the EU. Also, two vehicle classes (light and heavy duty vehicles [LDVs and HDVs]) were selected to provide LCA results on per-km-travelled basis. To this end, this study uses the Greenhouse gasses, Regulated Emissions, and Energy use in Transportation (GREET®) model [23], which was developed by ANL with the support of several programs in Department of Energy. GREET is structured to systematically examine the life-cycle energy use and emissions including GHG emissions and critical air pollutants associated with a wide range of fuel production pathways from various feedstock sources and vehicle technologies.

SYSTEM BOUNDARY AND APPROACH

System Boundary of WTW Analysis

Figure 1 presents the system boundary of the five DME pathways investigated in this study. The end of cycle is vehicle operation since DME is used as a transportation fuel. The LCA of transportation fuel is often called well-to-wheels (WTW) analysis. A WTW pathway consists of a well-to-pump (WTP) stage - covering the recovery and transport of feedstock, and the production, transportation and distribution of fuel - followed by a pump-to-wheels (PTW) stage representing vehicle operation. Note that, if there is co-produced steam, it is assumed to displace NG, and the energy and emissions associated with the displaced NG are taken into account as credit.

In Figure 1, the small- and large-scale DME productions represent around 25 MTPD (metric ton per day) and 3,600 MTPD, respectively. For NG, there are two options: one with integrated conversion from NG to DME via methanol (Case NG), and the other with two separate plants from NG to methanol followed by a methanol to DME production process (Case MeOH). It can be determined by considering the distances between the NG fields and the DME markets, and transportation infrastructure. NG would be transported to DME plants located near the demand where pipelines are well developed between them. If not, DME plants would be located near NG fields so that it could be converted to methanol for long-distance transport.

Biogas is a mixture of CH₄ and CO₂ generated from anaerobic digestion (AD) of organic wastes. Two types of biogas - LFG from landfilled organic wastes and manure-based biogas - were investigated for DME production as renewable alternatives. In Figure 1, these pathways are denoted as Case LFG and Case MANR, respectively. Since these resources are distributed sparsely in small volumes, it is assumed that DME is produced in small-scale plants that are co-located with the source of biogas.

Biogas is a waste feedstock, which would need to be treated if it were not diverted for energy recovery. For example, when LFG is collected, it is flared to reduce global warming potential of direct methane venting, or combusted to generate electricity. Similarly, manure needs to be treated by various processes (e.g., anaerobic lagoon, pit storage, soil application, etc.). During manure treatment processes, a large amount of GHG emissions (CH₄ and CO₂) can be generated. These waste treatment processes that would happen otherwise are the so-called counterfactual scenarios. This study defines business-as-usual waste treatment practices as the counterfactual scenarios. Specifically, biogas flaring is assumed for the counterfactual scenario of LFG while the current mix of manure treatment practices is selected for MANR. The amount of waste diverted for the fuel production would not require corresponding waste treatment in the counterfactual scenarios, avoiding energy uses and emissions associated with the waste treatment. Thus, these avoided energy uses and emissions in the counterfactual scenarios are taken into account as energy and emission credits for the WTW analysis of waste-based fuels.

Black liquor is produced as a byproduct of the Kraft process, the most dominant pulping process. Fibers in the wood are used to produce pulp, and materials other than fibers in the wood are dissolved in chemicals, which is called black liquor. Nearly all black liquor produced in modern pulp mills is recovered in a recovery boiler to support heat and power needs for the mill and to reduce environmental impacts [24]. Alternatively, black liquor can be used as a biofuel feedstock due to its high organic content. In this study, black liquor is assumed to be gasified to produce DME (Case BL) instead of being burnt in a recovery boiler. This black liquor-based DME production can be handled in two approaches: 1) treating black liquor as a burden-free renewable feedstock and 2) estimating incremental changes in a pulp mill by DME production. In the first approach, the system boundary includes the DME plant converting black liquor to DME, and the inputs and outputs of the DME plant are taken into account. Thus, all energy streams between the DME plant and the pulp mill are considered as inputs (if from the pump mill; e.g., black liquor) or outputs (if from the DME plant; e.g., steam and electricity) of the DME plant. On the other hand, the second approach assumes that black liquor-based DME uses a feedstock that is currently used for other applications (i.e., power generation), and calculates the incremental changes in the pulp mill as DME production was implemented to the reference plant with a recovery boiler. Thus, energy streams between the DME plant and the pulp mill are largely netted out. This study uses the second approach because most modern pulp mills currently use recovery boilers. Note that, since the incremental energy inputs depend not only on the alternative methanol production assumptions but also on the reference plants' performance, the second approach is subject to large variations and uncertainties.

All five cases are analyzed for the US and EU with different conditions such as NG recovery efficiencies, transportation methods and distances, manure shares, and electricity generation mixes. For EU, however, the integrated NG-to-DME conversion pathway is excluded since high NG price and the significantly long NG transmission via pipelines makes this pathway economically and environmentally less competitive. Detailed parameters associated with each process are represented in the following sections.

KEY PARAMETRIC ASSUMPTIONS OF WTW ANALYSIS

NG Recovery/Processing and Methanol Production

Key parameters for NG recovery/processing and methanol production are documented in Table 1. The assumptions for the US cases are from Burnham et al. [25,26] while those for the EU cases are based on the JEC WTT study [20]. The overall efficiency in the EU is estimated to be higher than that in the US. Also, CH₄ leakage rates and CO₂ venting estimates in the EU are generally lower than the US estimates. As a result of these key inputs, the EU's NG GHG intensity in this study is marginally lower than that of the US. Table 1 also includes the material and energy inputs, GHG emissions and critical air pollutant emissions for the methanol production pathway provided by Haldor Topsøe A/S. It requires mostly NG for both methanol production feed and process fuels while a small amount of electricity is purchased from the electric grid. It is assumed that coproduced steam displaces NG-derived steam with an 80% energy efficient boiler, which means 0.11 MJ of steam co-produced with 1 MJ of methanol is assumed to displace 0.14 MJ of NG.

Table 1. Key parameters for fossil NG recovery/processing and methanol production

	US [†]	EU
NG recovery and processing		
Data source	GREET 2015 [25,26]	JEC 2014 [20]
Overall efficiency	95%	98%
CH ₄ leakage rate (g/MJ)	0.09	0.08
CO ₂ venting (g/MJ)	0.77	0.54
GHG intensity (gCO₂e/MJ)	7.6	4.3
Methanol production		
Feed NG input (MJ/MJ)	1.45	
Process fuel NG input (MJ/MJ)	0.013	
Electricity input (MJ/MJ)	0.040	
Steam co-product (MJ/MJ)	0.11	
Emissions (grams/GJ)		
VOC	0.63	
CO	1.13	
NO _x	0.74	
SO _x	0.10	
CO ₂	13,200	

[†]US data includes both conventional NG and shale gas considering the production share

Biogas Recovery/Processing and Counterfactual Scenario Definition

Once LFG is generated from landfills, it needs to be upgraded to be used for DME production, and the detail processes and assumptions are presented in Mintz et al. [27]. Since the DME production process works better with around 28% CO₂ content a 1-stage cleanup process is preferred, and the 2nd cleanup process typically used to produce pipeline-quality NG is not required. Thus, the overall LFG cleanup efficiency is assumed at 92% while CH₄ leakage during this process is assumed at 1% by volume. The required energy for the NG processing is supported by a microturbine combined heat and power (CHP) generator using LFG.

For manure-based biogas, appropriate manure shares should be considered as CH₄ generation varies by manure type as represented in the study by Han et al. [28]. Table 2 shows manure shares and corresponding theoretical maximum CH₄ production in the US and the EU. The manure shares for the US and the EU have been collected from US EPA AgSTAR [29] and Agro Business Park [30], respectively. Theoretical maximum CH₄ production depends on manure types and climate conditions, and the data for the US and the EU are based on US Department of Agriculture (USDA) [31] and Intergovernmental Panel on Climate Change (IPCC) [32], respectively. It is assumed that manure is transported by heavy-duty truck for 5 km to AD for biogas production. By default, a mixed plug flow AD is used, and the biogas generated from AD is estimated using methane conversion factors (MCFs) which represent the ratio between the CH₄ generation and its theoretical maximum. Heat and electricity required to support AD have been calculated in [28]. Also, energy and emissions associated with the soil application of AD residue have been taken into account. Regarding the biogas upgrading processes, the same assumptions as in Case LFG have been used.

Table 2. Manure shares and theoretical maximum CH₄ production in the US and the EU

	Manure Share [29,30]		CH ₄ Max (m ³ CH ₄ /kg VS) [31,32]	
	US	EU	US	EU
Dairy	80%	18%	0.24	0.24
Other Cattle	10%	18%	0.33	0.18
Swine	10%	64%	0.48	0.45

As mentioned earlier, the avoided energy and emission burdens associated with the counterfactual scenarios should be taken into account. Note that the upstream stages that are common in the counterfactual scenarios and alternative DME production cases (including waste collection, uncontrolled emissions, etc.) are not included because these stages would happen regardless of the fuel development. The counterfactual scenarios differ by feedstocks. For LFG, flaring emissions can be calculated using the emissions factors developed in Mintz et al. [27]. For manure, Han et al. [28] provided energy and emissions associated with the counterfactual scenario based on the process shares, CH₄ production rates, and operating conditions of current manure treatment practices.

Methanol Production from Black Liquor

Methanol production via black liquor gasification has been analyzed using the data from Andersson et al. [33], which is presented in Table 3. Table 3 also presents additional cases from Ekblom et al. [24]. It can be found that methanol production increases demands for electricity and forest residue, which is caused by the loss of heat and power production in the pulp mills with the recovery boiler. Note that the baseline assumption for electricity need in Case BL is biomass-based electricity while the use of regional electricity generation mixes is also examined as a sensitivity case. Also, the plants in Andersson et al. [33] and Ekblom et al. [24] generate methanol with less than 0.1% water, which requires a lot of heat for distillation. However, when a DME process is attached to a methanol plant, distillation is typically not needed for the raw methanol. To the best of our knowledge,