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Review of Well-to-Wheel lifecycle emissions of liquefied natural gas heavy goods vehicles

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Abstract: It has been suggested that using liquefied natural gas as a fuel source for heavy goods vehicles could provide a reduction in greenhouse gas emissions. Various studies have investigated different aspects of the lifecycle emissions of natural gas heavy goods vehicles throughout the past decade, however, there has been little comparative analysis across these studies. This review provides a comprehensive examination of the well-to-wheel lifecycle emissions of liquefied natural gas for heavy goods vehicles in comparison to diesel, the current standard. A systematic selection criteria based on relevance to the defined well-to-wheel system boundary of liquefied natural gas as a fuel source for heavy goods vehicles, including greenhouse gas emissions, were augmented by the authors knowledge of the field. The various data are categorised by engine technology and model year (pre- and post-2015), average speed of the duty cycle, and then statistically analysed to identify clear trends and correlations in the emissions produced. The two primary factors affecting the well-to-wheel greenhouse gas emissions of natural gas heavy hoods vehicles are: (i) natural gas engine fuel efficiency relative to diesel, and (ii) methane leakage across the supply chain. Methane leakage rates are a significant uncertainty and range from 0.3-20% of throughput. With long-term perspective of efficiency penalty (10%) in natural gas engines, the well-to-wheel greenhouse gas emissions reduction of natural gas fuelled trucks against diesel is up to 10%, which appears insufficient toward net zero emissions by 2050. The use of biomethane further reduce the greenhouse gas emissions by 34-66% depending on the engine technology. Controlling fugitive methane emissions in the fuel production and supply chain remains critical.

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1 Introduction

Greenhouse gas (GHG) emissions from the transport sector have increased at a faster rate than any other energy-consuming sector [1], and in 2016 the transport sector was responsible for 24% of global CO_2 emissions [2]. The European Union announced that emissions from domestic transport increased by 0.8% over 2018-2019 and dropped by 12.7% in 2020 due to the drastic decrease in transport activity during the Covid-19 pandemic [3]. The reported global transport emission is 7.2 Gt CO_2 in 2020, down from nearly 8.5 Gt CO_2 in 2019 [4]. Road freight in isolation accounts for 7% of global CO_2 emissions and over the period 2000-2015 CO_2 emissions attributable to road freight increased by 2.8% per year and contributed to >40% of the growth in the transport sectors CO_2 emissions [5]. The contribution of road freight to total emissions varies by region: 90% of the increase in CO_2 emissions over this period was in emerging economies led by China (approximately 25%) [5]. In industrialised economies, road freight emissions are not reducing in line with other parts of the transport sector; for example, in the UK, despite progress in reducing GHG emissions produced by the transport sector during the period 2003-2015, GHG emissions from heavy goods vehicles (HGVs) decreased by only 2.7%, significantly less than the reductions for light vans (20.7%), buses and coaches (24.2%), and cars and taxis (10.2%) [6], and have been increasing in the years since [7].

The emissions produced by HGVs are also relevant in the context of urban transport, where noxious emissions can have detrimental impacts on public health. Approximately 54% of the world's population lived in urban areas in 2014, and this figure is expected to rise to 60% by 2030 and up to 66% by 2050 [8]. HGVs contribute to reduced air quality due to the emission of a range of air pollutants, including nitrogen oxides (NO_x), particulate matter (PM), carbon monoxide (CO) and unburned hydrocarbons. In 2015, heavy duty vehicles (HDVs) were responsible for more than 40% of NO_x emissions and 50% of $PM_{2.5}$ emissions from the transport sector globally [9]. Alternative fuel sources offer the potential to decarbonise and reduce oil dependency in the road freight sector. However, they currently account for only 3.4% of final energy in road freight transport (2.2% biofuels and 1.2% natural gas) [5]. Natural gas has been considered as an alternative to diesel for HGVs for a variety of reasons, including: energy security [10], economics [11], operating noise reduction [12], and the potential to reduce emissions of GHGs and air pollutants [13].

There is a significant variation in the types of vehicles used for road freight, with different vehicles used for specific applications across a wide range of weight categories. HGVs are typically defined as commercial vehicles with a gross vehicle weight greater than 15 t, serving long-haul routes, having two to four (or more) axles, and a power rating between 200-600 kW. HGVs account for approximately 70% of freight activity and about 50% of truck energy use [5]. Statistics for the European Union (EU) indicate the dominance of long-haul freight transport by HGVs: more than 90% of freight tonne-kilometres were completed using vehicles with a maximum gross vehicle weight exceeding 20 t [14] and 78% of tonne-kilometres are from trip distances over 150 km [15]. HGVs are a subgroup of HDVs, which also includes other types of heavy vehicles that are used for purposes other than transporting goods. The vast majority of HGVs (96.5%) use diesel according to the EU market share in 2020 [16]. The other types of freight vehicles are beyond the scope of this study. The comprehensive studies on all freight vehicles can be found in Speirs et al. [17]. Natural gas is a mixture of paraffinic hydrocarbons such as methane, ethane, propane, and butane. Small amounts of heavier hydrocarbons, such as ethylene, may be present and trace amounts of hydrogen sulphide and nitrogen may also be present. The energy density (per unit weight and volume) for several transportation fuels, including liquefied natural gas (LNG) and compressed natural gas (CNG), are shown in Figure 1. The energy content of natural gas (CNG or LNG) per unit weight is approximately 15% higher than diesel fuel (using typical net calorific values of 50 MJ/kg and 43 MJ/kg for natural gas and diesel, respectively) [18], indicating that natural gas can offer the same amount of energy for less weight. However, natural gas has a significantly lower density than diesel; at atmospheric temperature and pressure the density is approximately 1,000 times lower than diesel. Natural gas must be either compressed to a pressure of 200-300 bar (CNG) or liquefied by cooling it to -162°C (LNG) to increase the volumetric energy density so that it can be stored on vehicles in on-board cylinders [19]. LNG is approximately 600 times more dense than natural gas at atmospheric temperature and pressure, whereas CNG is only 200-300 times denser. This means that LNG can offer 2-3 times the energy for the same capacity fuel tank, directly translating to 2-3 times greater vehicle range. However, the energy content of LNG per unit volume is still below diesel, meaning that LNG storage tanks would take up more space on the vehicle.

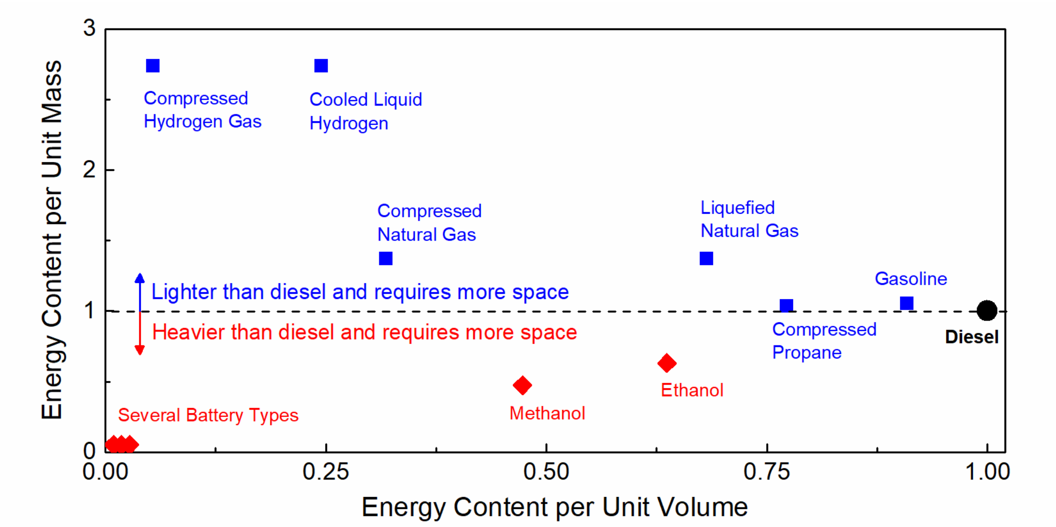

Figure 1: Comparison of the energy density of various transport fuels relative to gasoline. Data from [20].

Lifecycle assessment allows for a comprehensive understanding of environmental impacts, including climate change, air quality, and human health. Many studies have investigated the well-to-wheel lifecycle emissions of NG HGVs in comparison to diesel or gasoline. These studies employed lifecycle assessment modelling tools such as GREET [21-23], EMFAC [21], SimaPro [24, 25], Simcenter Amesim [26] or ad-hoc software developed by the vehicle manufacturer [27, 28] or type-approved data for conventional vehicles and real-world emission data [13, 29]. These emission factors apply general assumptions and ignore specific conditions such as driving regime and road type. To the best of the authors' knowledge, only Clark et al. [30] conducted a pump-to-wheels analysis from the US heavy-duty transport sector using real driving emissions. The authors characterised the tank-to-wheel emissions by measuring twenty-two natural gas fuelled transit buses, refuse trucks, and over-the-road tractors. Although the real-world emission factors are essential to estimate more realistic lifecycle assessment, study employing practical data is still lacking.

The primary objective of this review is to evaluate the state-of-the-art literature to ascertain the potential for LNG to reduce lifecycle greenhouse gas emissions produced by road freight. We consulted measured in-use emissions data including real-driving emission data and categorised them by the engine technologies and model year/emission standard, and average speed categories representing the different operational types of urban, rural/regional, and long-haul, respectively. As discussed above, HGVs operating on long-distance routes are responsible for the majority of freight activity. We therefore focus this review on HGVs, vehicles with a gross vehicle weight greater than 15 t. Where evidence from other types of vehicles is used, this is explicitly highlighted. For perspective, reference is made to diesel HGVs, as this is the dominant fuel type. We consider the full well-to-wheel (WTW) lifecycle, comprising the fuel supply chain (well-to-pump, WTP), refuelling (pump-to-tank, PTT), and vehicle operation (tank-to-wheel, TTW). Our review is focussed on LNG given its higher range capabilities compared to CNG. CNG pathways are different in the WTP and PTT phases, and these are out of scope, however natural gas engine technologies are able to operate with either CNG or LNG. Therefore, when in our review of the TTW phase, we include data on CNG heavy vehicles. Biomethane has the potential to significantly reduce WTP GHG emissions. However, we note that there are a range of different feedstocks and production pathways that contribute to a wide range of WTP emissions estimates [31, 32]. A complete evaluation of these different pathways is out of the scope of this review but in our Synthesis of the evidence (§7), we quantify the WTP emissions reductions that are required to reduce total lifecycle (WTP + PTT + TTW) emissions relative to diesel trucks.

The remainder of this article is structured as follows: Section 2 describes the systematic selection criteria used to identify relevant articles for this review. Section 3 provides an overall WTW systems analysis to identify the factors that influence the ability of LNG to be an environmentally viable alternative fuel for HGVs. Section 4 examines the WTP phase of the LNG supply chain, and the range of methane emissions produced during this stage. Section 5 investigates the mechanisms and ranges of methane leakage during the storage and refuelling phases (PTT stage). Section 6 focuses on the TTW, which assesses the literature on the various natural gas engine technologies, GHG emissions produced (CO_2 and CH_4), and the air pollutants that affect air quality. An evaluation of the literature surrounding current and predicted future fuel efficiency of natural gas HGVs is also presented. Finally, Section 7 presents a synthesis of the literature review and identifies where current and future natural gas engines need to be in terms of fuel efficiency and emissions compared to diesel engines, with consideration of biomethane.

2 Systematic Selection Criteria for Article Search

The articles selected for the review follow a systematic selection criteria based on relevance to the defined WTW system boundary of LNG as a fuel source for HGVs. The selection of relevant articles is based on the authors' knowledge of current and previous research projects related to low carbon vehicle technologies, air quality, sustainable transport, and knowledge of the surrounding sectors. In addition to work that was known to the authors, a systematic review of the peer-reviewed literature was undertaken through an online search of various databases including ScienceDirect, Scopus, Web of Science, and Google Scholar using Boolean combinations of the search terms listed in Table 1. Where grey literature is included, it is done so to incorporate the most recent empirical evidence and these reports are only included if empirical data is quoted directly and includes the experimental methodology and conditions.

The data that we have gathered as part of this review is available in the Supporting Information.

Table 1: Search terms used in the systematic literature review.

Vehicle	Emissions	Fuel	Modelling	Geographical Areas
Heavy Goods Vehicles	NOx	Methane	Lifecycle	US
HGV	SOx	Natural gas	Whole systems	"United States"
Heavy duty vehicles	Particulate	LNG	GREET	EU
Heavy duty engine	PM	CNG		"European Union"
Low carbon vehicle	Greenhouse Gas	Hydrogen		UK
LCV	GHG	Battery electric		"United Kingdom"
Zero Emissions Vehicle	Carbon Dioxide	Fossil gas		"The Netherlands"
ZEV	CO_2			Japan
Transport	Methane slip			Korea
	Particle Number			Germany
	PN			China
	SPN			World
	Methane			Global
	CH_4			

3 Natural Gas Heavy Goods Vehicles

In 2019, it was reported that there were over 28 million natural gas vehicles and over 33,000 refuelling stations across the world, with over 70% of vehicles in the Asia-Pacific region [33]. The majority of these vehicles are not freight vehicles, with trucks accounting for about 1% of total stock in 2015 [5]. In the United States, the use of natural gas trucks became more attractive with the expansion of domestic shale and tight gas production, leading to a dramatic drop in wellhead natural gas prices from 2009 [5]. The Fixing America's Surface Transportation Act, which requires the United States Department of Transportation to set aspirational targets for the deployment of infrastructure for alternative fuels along key corridors, has promoted the development of natural gas stations since 2015 such that there are 1,680 CNG stations and 144 LNG stations in 2022 [34]. In the North American market, several natural gas HGV models are offered from different original equipment manufacturers.

The market growth for natural gas trucks in China has been driven by several factors including the favourable price differential to diesel, the low cost of retrofitting existing vehicles to run on CNG, and government policies aimed at improving air quality. The use of natural gas in transport has increased by an annual growth rate of approximately 11% between 2010 and 2016, of which a significant share is attributed to natural gas trucks [5]. The number of stations supplying natural gas in China has grown from around 1,000 in 2008 to 7,950 in 2016, and the number of LNG heavy-duty vehicles grew from 7,000 in 2010 to 132,000 in 2015 [5]. The total number of natural gas heavy duty trucks was estimated to have reached 325,000 in 2017 [35].

In the EU, there are approximately 9,350 medium- and heavy-duty natural gas trucks, with over 80% of these trucks operating in Italy, Sweden, Spain, and France [36]. Compared to China and the United States, there is a reduced cost benefit of natural gas and fewer government incentives have been offered. However, the Alternative Fuels Infrastructure Directive requires EU member states to develop national policy frameworks to promote and develop the relevant infrastructure for alternative fuels including CNG and LNG. The directive suggests that the average distance between refuelling stations should be 150 km and 400 km for CNG and LNG, respectively [37].

Natural gas has a lower CO_2 intensity than diesel [38]; the principal component of natural gas, methane (CH_4), has a higher ratio of hydrogen to carbon atoms (4:1) than the average for diesel (~2:1), and therefore less CO_2 is emitted per unit of chemical energy released by combustion [39]. However, the lower carbon content of natural gas compared to diesel does not necessarily result in reduced GHG emissions across the WTW lifecycle once engine efficiency and methane leakages are taken into account. The global warming potential (GWP) of fossil methane is 29.8 ± 11 and 82.5 ± 25.8 times that for CO_2 for 100- and 20-year time horizons, respectively [40]. Therefore, any methane leakage can offset the lower carbon intensity of natural gas. Methane leakages can occur throughout the fuel supply chain and operation of the vehicle and it is therefore vital to consider GHG emissions across the entire WTW lifecycle [5].

4 Well-to-Wheel Systems Analysis

4.1 Previous studies

The entire WTW lifecycle of LNG as a vehicle fuel is outlined in Figure 2, beginning with the production, transmission, and storage of the natural gas, further processing and transport to refuelling stations, operations at fuelling stations, and ending with the vehicle operation [30]. The methane leakage rate across the WTW lifecycle is the principal factor for evaluating any potential environmental benefits to using natural gas as an alternative fuel source. It should be noted that embodied emissions are considered out of the scope, as for high-mileage HGVs the operational emissions are likely to dominate.

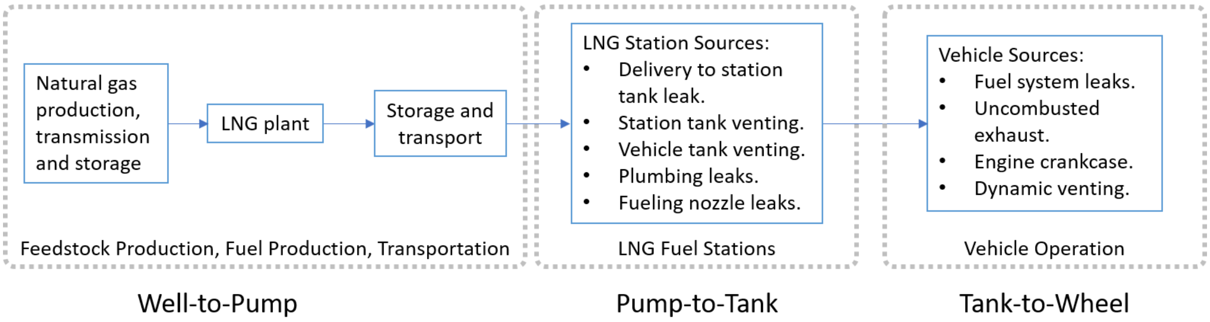

Figure 2: The well-to-wheel lifecycle of liquefied natural gas as a transport fuel with the system boundaries. Note: embedded/embodied emissions are out of scope.

In a recent whole-lifecycle evaluation of natural gas vehicles, Cai et al. [23] presented a comparison of the WTW CO_2-equivalent (CO_2e) emissions for LNG and CNG HGVs. The study found that the WTW GHG emissions of the natural gas vehicles were 1%, and 6% higher than an equivalent diesel vehicle, respectively. The WTW GHG emissions of the vehicles were strongly dependent on the vehicle fuel economy, and the results reflect that natural gas engines are currently not as fuel efficient as diesel engines - 76-77% of the WTW GHG emissions arose from tailpipe CO_2 emissions for the natural gas vehicles. The supply chain contributed 19% of the WTW GHG emissions for the LNG short-haul truck and approximately 20% for the CNG vehicles. The difference between the LNG and CNG pathways was attributed to higher rates of methane leakage from long-distance and local transmission of natural gas in the CNG pathway, whereas LNG is typically transported with lower rates of methane leakage. For the LNG pathway, the liquefaction process and methane leakages due to LNG boil-off were identified as the main contributors to GHG emissions in the supply chain stage.

This is in line with two studies focused on the UK and Swedish markets that found that, while the vehicle operation has a significant impact on any potential emissions reductions, the WTT stages can also provide a substantial contribution if there are methane leakages throughout the supply chain [41, 42]. From these studies examining the WTW lifecycle, it is clear that there are two principal factors that influence the ability of natural gas vehicles to reduce GHG emissions [5, 23]:

1. The vehicle's fuel efficiency relative to equivalent diesel vehicles.
2. The methane leakage rate including:
    a. across the supply chain (WTP),
    b. at storage at refuelling stations and during refuelling (PTT), and
    c. during the operation of the vehicle (TTW).

Cai et al. [23] presented a sensitivity analysis examining various fuel economies and WTW methane leakage rates. The analysis shows that the methane leakage rate across the entire WTW lifecycle must be less than 2.8% (relative to throughput) for LNG vehicles to provide GHG emissions reductions [23]. For the North American supply chain, Cai et al. [23] stated it is possible if improvements are made to vehicle technologies and methane leakage is reduced throughout the supply chain. However, if methane leakages across the supply chain are larger than current estimates suggest, then the fuel economy for natural gas vehicles would need to significantly improve to be 'on par' with diesel [23].

Cooper et al. [22] conducted an environmental life cycle assessment considering CNG, LNG (dedicated and dual fuel), diesel, biodiesel, dimethyl ether and electric battery as fuels for HGVs. The results indicate that while natural gas offers benefits over diesel in all environmental indicators considered, the magnitude of the reductions is not enough for the UK to meet medium-long term climate change targets and limits for some air pollutants are exceeded. The conclusion of this study is in line with the findings from recent studies claiming little or no climate benefits from natural gas trucks [35, 43-45].

4.2 Regional Differences

One important factor to consider across the WTW lifecycle is the LNG supply chain, which will vary depending on the region. For instance, some regions have greater international transportation distances (e.g. from North or West Africa to Asia), as well as different levels of leakage in high-pressure pipelines for national distribution. Regions also have different approaches for when LNG is processed. Arteconi et al. [21] presented a case study comparing the lifecycle GHG emissions for diesel and LNG HGVs in Italy. Two alternative LNG pathways were investigated: 1) LNG is imported into Europe via LNG methane carriers to regasification terminals (LNG-TER), and 2) LNG is produced locally in small-scale liquefaction plants at service stations (LNG-SSL). Their results are summarised in Table 2 and show that importing LNG and using a regasification terminal (LNG-TER) resulted in a 10% reduction in total WTW GHG emissions compared to an equivalent diesel vehicle. However, using small-scale regasification plants (LNG-SSL) was only found to provide a 3% reduction in GHG emissions compared to diesel. The lower tailpipe emissions were offset by greater GHG emissions in the supply chain as a result of inefficient small-scale liquefaction systems. This does indicate that the total emissions of the LNG-SSL pathway would decrease linearly with an increase in the efficiency of liquefaction such that if the efficiency of small-scale plants were to reach 90%, the WTW emissions of this route would be 9% lower than diesel [21].

Table 2: Well-to-wheel heavy goods vehicle greenhouse gas emissions for two liquefied natural gas pathways (LNG-TER: regasification terminal, LNG-SSL: locally produced small-scale liquefaction at refuelling station) and a diesel baseline for comparison, from [21].

Pathway	Production [kg CO_2e/km]	Distribution [kg CO_2e/km]	Combustion [kg CO_2e/km]	Diesel pilot [kg CO_2e/km]	Total [kg CO_2e/km]
Diesel	0.200	0.021	1.635	–	1.856
LNG-TER	0.160	0.088	1.401	0.015	1.664
LNG-SSL	0.389	0.001	1.401	0.015	1.806

For North American natural gas supply chains, Cai et al. [23] found that, in terms of natural gas transmission, the CNG supply chain generally produces greater GHG emissions compared to LNG due to leakages along the long-distance pipelines used to deliver CNG to refuelling stations. Since LNG liquefaction plants are often located close to natural gas sources, transmission occurs through shorter high-pressure pipelines and can result in less methane leakage.

The following sections discuss each aspect of the WTW emissions of LNG as a fuel for HGVs in detail.

5 Well-to-Pump

The first stage of the LNG lifecycle examined is the WTP phase, which covers the extraction of the natural gas to its distribution to refuelling stations. This section is focused on a literature review to establish general supply chain emissions from natural gas, and an in-depth examination of the supply chain emissions for LNG as a transport fuel.

5.1 Supply Chain Emissions

The survey of WTP natural gas emissions presented herein builds upon the 2015 review undertaken by Balcombe et al. [46]. Several notable updates since the 2015 study include two studies by Balcombe et al. [47, 48] that incorporate new data from 2016 and 2017, respectively. One issue with the literature examining emissions across the WTP supply chain is the lack of standard reporting units and variation in estimation methods. There are also significant differences in natural gas supply chains in different regions.

Balcombe et al. [48] conducted a systematic literature review and reported the mean and estimated distributions of emissions at each stage in the natural gas supply chain from over 450 studies up to 2017. Other relevant recent studies include the work presented by Cai et al. [23], Burnham [49], Alvarez et al. [50], and Littlefield et al. [51]. The ranges of methane emissions presented in these studies vary significantly and the majority are focused on the supply chains associated with North American natural gas. The literature suggests that methane emissions (as a percentage of total volumetric throughout) are within the range of 0.2-10%. The high variation is primarily due to the different methodologies, observational data, and whether the estimates were made using a bottom-up (i.e. observation of methane leakages along each stage of the supply chain) or top-down approach (i.e. observations of aggregate natural gas supply and demand). Furthermore, the ranges are sensitive to the inclusion of super-emitters, which can be described as a small number of sites (e.g. extraction or procession plants) that produce extremely high methane emissions [23, 50]. The aggregated methane emissions for a typical natural gas supply chain (WTP) from the literature are given in Table 3.

IEA methane tracker [52] provides methane emissions estimates for 2021, which is 2.85% higher than the methane emissions for 2020 and 0.25% lower than that for 2019. The report illustrates that, following the COVID-induced decline in 2020, a year-on-year increase in energy-related methane emissions of almost 5% is largely due to higher fossil fuel demand and production as economies recovered from the shock of the pandemic.

Table 3: Aggregated ranges of methane emissions in the natural gas supply chain (from extraction to distribution).

Year	Region	Methane Leakage Percentage of Throughput [%]			Notes of Study	Source
		Lower	Central	Upper		
2020	North America		0.5*1		Bottom-up inventory based on pipeline leak measurements	[53]
2019	North America		1.0*1		Bottom-up inventory	[54]
2019	China	-	0.39 (2008) 0.57 (2016)	-	Literature survey and bottom-up accounting.	[55]
2018	Global	0.80	-	2.20	Literature survey and Monte Carlo simulation.	[48]
2018	North America	2.00	2.30	2.70	Facility-scale bottom-up validated with	[50]

					top-down studies using a 95% confidence interval.	
2017	Global	0.20	-	10.00	Literature survey.	[47]
2017	North America	1.32 (conventional gas supply)	-	1.34 (shale gas supply)	Literature survey and lifecycle analysis using GREET and EPA 2016 GHGI.	[23]
2017	North America	1.30	1.70	2.20	Bottom-up and Monte Carlo lifecycle analysis with 95% confidence interval.	[51]
2015	Global	0.20	2.20 *2 1.60 *3	10.00	Literature survey.	[46]
Summary	Global	0.20	1.30	10.00		

Note:*1 estimated values from He et al. [45], *2 mean estimate, *3 median estimate.

5.2 Source Specific Emissions

The overall methane leakage can be further separated into each stage of the supply chain. Cai et al. [23] presented a breakdown of the methane emissions across the North American natural gas supply chain from various sources and compared their estimates to earlier studies, shown in Table 4. It is evident from the table that the processing is the least significant stage in terms of methane leakage, while the leakage during production (25-57%), transmission and storage (19-32%) and distribution (10-32%) can all produce significant emissions. The WTP values presented in Cai et al. [23] relies on the assumptions used in GREET for natural gas North American pathways and are presented in Supplement Information, SI. 1.

Table 4: Natural gas supply chain emissions per stage across the well-to-pump phase, from [23].

Stage of the Supply Chain	Methane Emissions (Percentage of Volumetric Natural Gas Stage Throughput [%])							Range Contribution to Total	
	EPA GHGI 5 yr. Avg.	EPA GHGI 2011 Data	EPA GHGI 2012 Data	EPA GHGI 2013 Data	EPA GHGI 2014 Data	Cai et al. Conv. Gas (2016)	Cai et al. Shale Gas (2016)	Low	High
Gas Field	1.32	0.45	0.34	0.34	0.71	0.7	0.77	25%	57%
Processing	0.17	0.12	0.13	0.16	0.13	0.13	0.13	7%	12%
Transmission and Storage	0.49	0.44	0.39	0.44	0.36	0.36	0.36	19%	32%
Distribution	0.57	0.36	0.4	0.43	0.14	0.14	0.14	10%	32%
Total	2.53	1.36	1.25	1.36	1.33	1.32	1.34		

6 Pump-to-Tank

The PTT phase comprises the storage of LNG at the refuelling station to its delivery into the vehicle's tank. There is limited published literature on methane emissions during this phase, however, methane leakages can occur through several mechanisms at refuelling stations and may constitute up to 21% of the total pump-to-wheel (PTW) emissions [30]. Common to both LNG and CNG stations are continuous unintentional leaks from fuel nozzles (and other fuel delivery system components) due to imperfect seals that allow pressurised natural gas to escape into the atmosphere. Furthermore, methane emissions can occur during the hose-vehicle coupling at the start and end of each refilling event [30].

6.1 LNG Refuelling Stations and Processes

As LNG is a cryogenic liquid that is stored at temperatures as low as −162 °C, heat transfer from the environment to the stored LNG causes evaporation and the generation of boil-off gas (BOG). This causes a build-up of pressure within the storage vessels. To ensure that pressures remain within safe limits and the LNG pressure and temperatures are maintained within a suitable range for delivery to vehicles, the BOG must be vented, and in some station designs, the BOG is vented directly to the atmosphere [56].

LNG can be delivered to vehicles at a refuelling station in two forms: unsaturated LNG (dispensed at less than −143 °C and 0.34 MPa), or saturated LNG (dispensed at −125 to −131 °C and 0.69 to 0.93 MPa). Unsaturated LNG has a lower temperature, higher density, and can be stored on vehicles longer than saturated LNG. However, the lower pressure of the unsaturated LNG means that auxiliary equipment in the vehicle fuel supply system is required to increase the fuel pressure delivered to the engine [56]. There is a lack of evidence regarding fuel station emissions using these different forms of LNG.

For LNG refuelling in general, the BOG generated in the customer vehicle's storage tanks must be dealt with prior to refuelling, as otherwise there is not enough of a pressure difference between the pump system and the on-board storage tank. The operating sequence of refilling a vehicle's LNG tank at a refuelling station must manage the high-pressure BOG in the on-board LNG tanks. Several options for the refuelling operating sequence exist [56]:

1. *No vapour back to the station*

   These use the station pressure to overcome the tank pressure and condense the BOG (only possible if the on-board tank pressure has sufficient margin below the relief valve pressure and the station pump has sufficient discharge pressure available).

2. *Vapour back to the station*

   These use a vapour return line routed through the fill receptacle or a separate vapour return line to reduce the tank pressure prior to filling.

3. *Vapour back to the station and continue to vapour back during fill operation*

   The transfer of LNG from the refuelling station to an on-board LNG tank while the BOG in the tank is returned to the station by the vapour return line. In this method, the LNG pressure at the station does not need to be too high; however, it is only possible with a separate vent return line.

4. *Manual vent of BOG*

   Venting the BOG to the atmosphere to reduce the on-board tank pressure before proceeding with the process described in option (1).

The storage tanks at refuelling stations also require careful management to prevent the venting of methane into the atmosphere due to BOG generated as a result of heat transfer into the tanks. Most LNG stations are served by periodic deliveries from tanker trucks that refill the storage tanks, referred to as 'offloading'. There are three main methods of creating a pressure difference between the source tank and storage tank to drive refuelling [57]: (1) a pressure build-up unit (PBU), comprising an air-heated heat exchanger that evaporates LNG to create a high-pressure false head, (2) a pump, or (3) a combination of a PBU and pump. A vapour return pipe may be added to the system to decrease the pressure difference between the storage and source tanks. However, in systems relying only on a PBU, the BOG in the storage tank may need to be vented to atmosphere before refuelling can take place.

While it is possible to re-condense the BOG using an on-site liquefier or by directing the BOG into the low-pressure natural gas grid [56], these components significantly increase the cost of refuelling stations. A review of existing LNG station designs found that the majority of refuelling stations available on the market had no BOG management. Of patented LNG refuelling station designs, 44% were found to have no BOG management [56].

6.2 Methane Emissions at Refuelling Stations

The latest measurements of methane emissions at refuelling stations are presented by Clark et al. [30], and these data have been incorporated into the latest evaluation of WTW emissions by Cai et al. [23]. The study by Clark et al. [30] followed a bottom-up measurement methodology that measured methane emissions from different components of the refuelling station system. Sources of methane emissions included leaks from mechanical fittings at stations and on vehicles, vents from storage tanks, compressors and fuelling systems, and releases during fuelling hose disconnects, as well as vehicle tailpipe and crankcase vent emissions (covered in Section 5). Hand-held methane detectors were used to locate leaks from stations and on-board fuel storage and transfer systems. Methane emissions from refuelling stations were gathered from six LNG stations, all fed by cryogenic tanker truck deliveries.

The LNG stations visited were all commissioned after 2011. LNG storage tank boil-off, pressure relief valve venting and manual venting of on-board LNG vehicle tanks by drivers prior to refuelling were also observed during the study.

The ranges of methane emissions given by Clark et al. [30] for the various sources are given in Table 5. Regarding delivery of LNG, Clark et al. [30] measured an upper bound of 0.381%. However, in their modelling study Sharafian et al. [57] estimated that for stations equipped only with a PBU, methane emissions may be up to 4.9% and 10.4% of throughput, with and without a vapour return pipeline, respectively. While we would expect that most new LNG refuelling stations have BOG management in place [58], there is a lack of quantitative evidence on offloading processes at stations globally and these results indicate the potential for extremely high methane leakage rates.

Regarding venting from the vehicle fuel tank, UNECE Regulation 110 [59] requires that, after a full-fill, the minimum holding time without venting of LNG in vehicle fuel tanks be 5 days (Annex 3B, paragraph 2.7) [59]. For intensively used vehicles, it is unlikely that LNG will be vented from the fuel tank. However, in the event that an LNG tank remains full after 5 days, 2-4% of LNG may boil-off and be vented for every following day (Gunnarson et al. [60], Ursan et al. [61]). Therefore, the upper bound of vehicle fuel tank venting is estimated as 3% of throughput.

In summary, the main source of methane emissions in the PTT phase is the venting of methane to the atmosphere due to BOG pressure build-up in the station and on-board storage tanks. The management of BOG from on-board LNG tanks, the flexibility of stations to refuel vehicles with different fuel supply systems, and the minimisation of BOG generation in the station storage tanks and venting to the atmosphere during offloading are critical to reducing fugitive methane emissions. The maximum methane leakage rates in Table 5 represent the worst-case situation evidenced by literature and they serve to highlight that extremely high methane emissions are possible under certain circumstances.

Table 5: Summary of the range of methane leakage (as a percentage of throughput) during all stages of the pump-to-tank stage.

Source	Methane Leakage Percentage of Throughput [%]			Source
	Low	Middle	High	
Delivery (Offloading)	0.071	0.128	10.00	[30] and [57]
Station tank BOG	0.000	0.100	2.000	[30]
Continuous leaks at stations	0.000	0.010	0.040	[30]
Fuelling nozzle	0.000	0.015	0.279	[30]
Vehicle fuel tank	0.000	0.100	3.000	[30],[59],[60],[61]
Vehicle manual vent	0.000	0.100	4.200	[30]
Total	0.071	0.452	18.301	

7 Tank-to-Wheel

The TTW phase is focused on the emissions produced by the vehicle during operation. The following presents a description of the various natural gas engine technologies, including their fuel efficiency relative to diesel engines, GHG and air pollutant emissions. The engine technologies are identical for CNG and LNG, and so results from both fuels are included. In addition to the CO_2 emissions produced by the vehicles, preventing methane emissions is vital for a competitive natural gas HGV. The various mechanisms through which methane can be emitted during the TTW phase are also described in this section. The review of non-GHG air pollutant emissions is provided in Supporting Information, SI. 2.

Lifecycle inventory usually employ a simple correlation to estimate the vehicle emissions data, which result in large gaps against the real emissions in practice. The use of in-use real driving emissions data greatly reduces the uncertainty in the evaluation of TTW GHG emissions. However, there are still high variability in the real driving emissions depending on the vehicle type, the use of after-treatment system, driving habits, and road conditions. The emissions data in this study are therefore categorised by the engine technologies and model year / emission standard, with two different categories: 1) before 2015/Euro V, and 2) after 2015 or Euro VI. We have chosen this distinction as heavy-duty emissions regulations in the US (US 2010 standards) and Europe became aligned with the introduction of Euro VI in 2013/14 [9] such that HGVs compliant with these standards employ the same pollutant emissions control technologies for PM and NOx, namely diesel particulate filters and selective catalytic reduction, respectively. Further classification of the data is performed by the average speed of the duty cycle. The emissions data are separated, where possible, into three average speed categories: 1) $v_{avg} \leq 30$ km/h, 2) $30 < v_{avg} \leq 60$ km/h, and 3) $v_{avg} > 60$ km/h. These ranges broadly represent the different operational types of urban, rural/regional, and long-haul, respectively.

We note that instead of taking the average speed of a given test or drive cycle, the EU's in-service conformity testing defines speed ranges of 0-50 km/h for urban, 50-75 km/h for rural and >75 km/h for motorway operation [62]. Vehicle emissions results derived by analysing emissions binned by instantaneous vehicle speed (e.g. [63]) may show different results compared to using the average speed of a duty cycle and these studies are therefore omitted from our analysis.

7.1 Natural Gas Engine Technologies

The use of natural gas (NG), gasoline, and diesel for transportation relies on internal combustion engines to convert the fuel's chemical energy into kinetic energy and vehicle motion. Many of the technologies used in natural gas engines are similar to conventional diesel/gasoline engines [64]. Internal combustion engines can be broadly classified into two categories; spark-ignition (SI), and compression ignition (CI), with similarities to gasoline and diesel engines, respectively. SI engines use a spark plug to ignite the air/fuel mixture within the cylinder, while the ignition in CI engines occurs solely due to the compression of the fuel. SI engines typically require near-stoichiometric[‡] fuel and air mixtures, while CI engines are able to operate in lean conditions, where there is more air than is required by the combustion process. The advantage of being able to run lean is that the intake air flow rate does not need to be throttled, as in an SI engine, and this provides a significant benefit in terms of energy efficiency [65-67].

Natural gas engines are either: 1) dedicated (mono-fuel) SI Otto-cycle engines, or 2) CI diesel-cycle engines that utilise a pilot injection of diesel for ignition, and so can be called dual-fuel engines [68]. For SI engines, the fuel octane rating is an important parameter to quantify the ability of the fuel to resist pre-ignition during compression (commonly known as knock). Natural gas has an octane rating of 120-130 [69], which is higher than typical gasoline at 90-98, therefore natural gas SI engines are

[‡] A stoichiometric mixture is the ideal air/fuel mixture that enables combustion to completely burn all fuel present in the cylinder.

able to operate with higher compression ratios and potentially be more energy efficient than gasoline SI engines. Diesel-cycle engines (CI) operate with natural gas-diesel mixtures. Due to the high octane rating of natural gas and low cetane number (a measure of the fuel's combustion speed) relative to diesel, it is not suitable for use in CI engines. Therefore, to utilise natural gas in CI engines, diesel is also injected into the cylinder to provide the ignition on compression. These CI engines are therefore called dual-fuel engines (distinct from bi-fuel engines which may be SI engines that operate on gasoline or natural gas independently). Depending on the engine configuration, between 0-95% of the fuel energy supplied to a dual-fuel engine may be in the form of natural gas, which is also referred to as the diesel substitution ratio [70].

The three common natural gas engine technologies used in HGVs are:

1) Spark-Ignition Stoichiometric (SIS)

A Spark-Ignition Stoichiometric (SIS) engine uses an exact air to fuel ratio required to combust all fuel molecules within the chamber. For natural gas, methane and air are completely converted to their products of reaction during combustion; H_2O, CO_2, and N_2. While these engines provide a cleaner combustion process and therefore cleaner exhaust gases compared to conventional engines, the fuel consumption is higher and power output lower relative to diesel engines [23].

2) High-Pressure Direct Injection (HPDI)

High-Pressure Direction Injection (HPDI) engines are a type of a dual fuel engine that use diesel as a pilot ignition source and injects the gas at high-pressure (e.g. >300 bar) into the combustion chamber at the end of the compression stroke. In HPDI engines, the diesel injection accounts for approximately 5% of the fuel energy, with the balance provided by natural gas [71]. Some studies have recently claimed that newer generation HPDI engines are able to offer similar levels of performance and drivability to diesel [51, 72].

3) Dual Fuel (DF)

Dual Fuel (DF) engines utilise two types of fuel to produce combustion as opposed to a single fuel source. Generally, diesel is the primary fuel and natural gas is added to the incoming air in the intake manifold and dual fuel engines are common as retrofitted diesel engines [39, 73]. This air/natural gas mixture is ignited by an injection of diesel at the end of the compression stroke. Dual fuel engines can offer advantages over other natural gas engine technologies, including higher thermal efficiency (relative to SI engines), flexible fuel capabilities (dual fuel engines can also run on only diesel), reduced fuel costs, along with the potential to reduce some air pollutant emissions [39, 74, 75].

An overview of the various engine technologies, after-treatment technologies, and fuel compositions are presented in Table 6. The following sections are focused on the literature examining GHG and air pollutant emissions produced by various natural gas HGVs and other heavy-duty vehicles.

Table 6: Natural gas engine technologies, after-treatment, and percentage of natural gas used, from [70, 72].

Engine Type	After-Treatment	Natural Gas Proportion Used [%]	Original Equipment Manufacturer
Spark-Ignition Stoichiometric (SIS)	3-way catalyst	100%	Cummins, Scania, Waukesha, IVECO
High-Pressure Direct Injection (HPDI)	Catalysed DPF, Urea SCR	95-98%	Westport, Volvo
Dual fuel (DF)	Oxidation catalyst	0-95%	Volvo (retrofit)

7.2 Tailpipe CO_2 Emissions

The tailpipe CO_2 emissions produced by various diesel and natural gas HGVs are examined in this section. Several on-road and laboratory studies have investigated the TTW GHG emissions produced by natural gas HGVs over different duty cycles and payloads [18, 39, 63, 65, 66, 76-84]. The summary of the CO_2 emissions produced by the various diesel and natural gas vehicles from the literature are presented in Figure 3 and Table 7. Tailpipe CO_2 emissions are a function of the CO_2 intensity of the fuel and the efficiency of the engine. Figure 3 indicates that there is significant variability in CO_2 emissions across all engine types and speed ranges reported in the literature.

An overall reduction in tailpipe CO_2 emissions is observed across most engine types comparing to pre-2015 and post-2015, with the exception of natural gas SIS engines for v_{avg} < 30 km/h, highlighting the progress made in reducing CO_2 emissions across different engine types. There is limited available data for natural gas DF engines since 2015. Regarding median post-2015 results, the HPDI engines produce the lowest median CO_2 emissions on regional/rural and long-haul duty cycles, with reductions of 27-30% compared to diesel. SIS engines achieve 20% and 12% reductions compared to diesel on regional/rural and long-haul duty cycles, respectively. However, HPDI and SIS engines have 8% and 19% higher CO_2 emissions on urban duty cycles, respectively. The results indicate that SIS and HPDI natural gas engines can provide significant reductions in tailpipe CO_2 emissions across regional/rural and long-haul duty cycles. The greater reductions for HPDI engines compared to SIS is due to the improved thermal efficiency of CI engines.

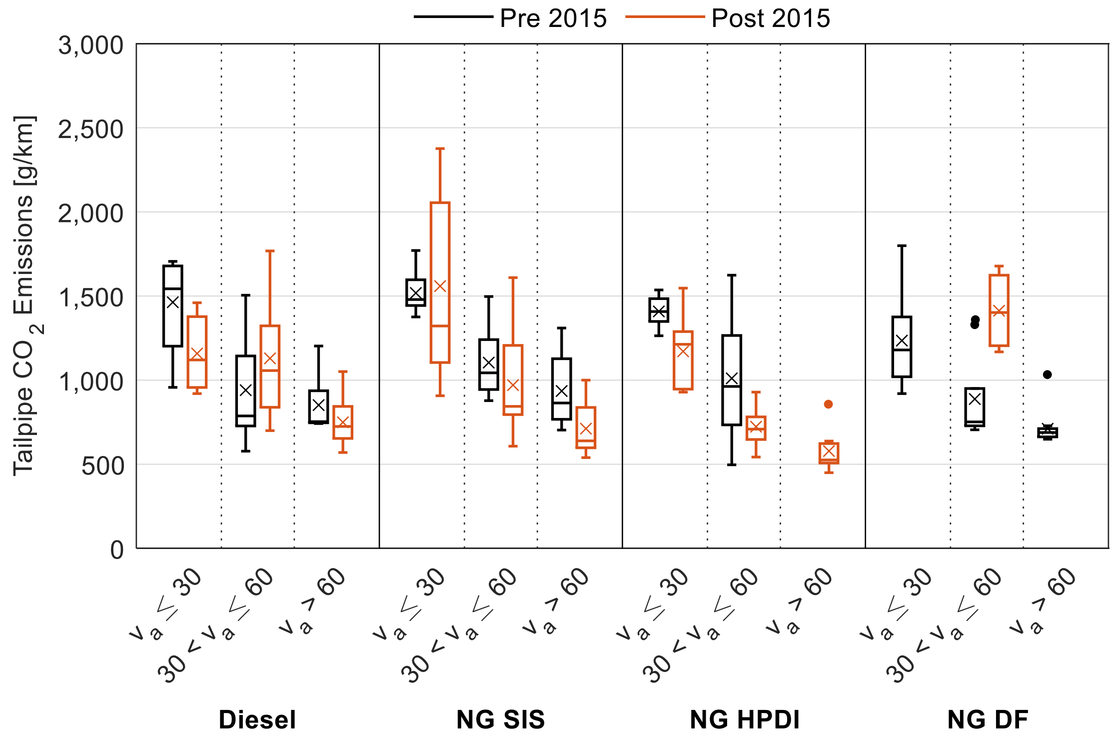

Figure 3: The distribution of tailpipe CO_2 emissions from diesel and natural gas engines pre- and post-2015 and for different average speeds.

Table 7: Summary of tailpipe CO_2 emissions from diesel and natural gas engines pre- and post-2015 and for different average speeds.

Model Year	Fuel Type	Engine Type	CO_2 Emissions [g/km]												Source
			$v_{avg} \leq 30$ km/h				$30 < v_{avg} \leq 60$ km/h				$v_{avg} > 60$ km/h				
			Mean	Med.	Q1	Q3	Mean	Med.	Q1	Q3	Mean	Med.	Q1	Q3	
Pre 2015 / Euro V	Diesel	CI	1,463	1,549	1,336	1,679	940	798	748	1,104	852	754	749	873	[39, 76-78]
	NG	SIS	1,518	1,479	1,451	1,565	1,104	1,044	963	1,175	936	864	819	981	[77-79]
	Dual	HPDI	1,409	1,408	1,367	1,464	1,011	963	832	1,249	-	-	-	-	[76, 77]
	Dual	DF	1,235	1,180	1,098	1,322	966	815	741	1,208	716	692	668	718	[39, 79, 82]
Post 2015 / Euro VI	Diesel	CI	1,158	1,120	970	1,338	1,130	1,057	8,66	1,311	750	725	656	843	[82, 83]
	NG	SIS	1,559	1,333	1,187	1,961	971	844	809	1167	712	639	600	799	[80-83]
	Dual	HPDI	1,171	1,213	952	1,274	723	734	662	760	579	526	515	594	[84]

	Dual	DF	-	-	-	-	1,467	1,467[1]	1,362	1,573	806[2]	[82]

1. Two data points so the median is equal to the mean; 2. One data point, therefore not included in Figure 3.

7.3 Methane Emissions

Methane slip occurs due to natural gas leakages throughout the vehicle system, of which there are three potential mechanisms. The first is the potential for unburned methane to be emitted via the tailpipe due to incomplete combustion. Generally, catalysts are used to control the tailpipe methane emissions. A three-way catalyst is paired with SIS engines, and an oxidation catalyst is used to control the emissions from CI engines [70, 85]. The second mechanism of methane leakage is through the engine crankcase. Methane can escape from the combustion chamber into the engine crankcase. If the engine crankcase is vented to the atmosphere, any methane present will also be vented. Crankcase ventilation systems and improved oxidation catalysts are currently available to minimise or eliminate crankcase methane emissions, however, up to 2015 at least, there has been little incentive for manufacturers to implement these technologies [86]. The final mechanism, limited to HPDI engines, is dynamic venting that occurs due to the behaviour (transient operation) of the fuel rail pressure control system and can emit small amounts of gas to the atmosphere via a pipe [86]. This section presents the findings of various studies investigating the methane emissions from three potential mechanisms; via the tailpipe, crankcase, and dynamic venting.

7.3.1 Tailpipe Methane Emissions

The range of tailpipe methane emissions produced by various natural gas vehicles (and one diesel for comparison) are presented in Figure 4 and Table 8.

Progress in reducing tailpipe methane emissions is observed for both the SIS and HPDI engines over time, however, it is clear that the lowest fuel-specific methane emissions are produced by SIS engines. The methane emissions due to stoichiometric combustion with a three-way catalyst are substantially lower due to the high exhaust temperature. There is insufficient recent data (post 2015) on DF engines. No significant trend is observed for the SIS on lower tailpipe methane emissions depending on the average speed of the duty cycle, while for the HPDI engines, show a decrease in the methane emissions as the average speed of the duty cycle increases. There are numerous factors which can explain the variation in methane emissions produced, from differences in vehicle age, catalyst temperature, engine speed, vehicle load, transient behaviour, and emissions diffusion between neighbouring micro-trips [30]. These results suggest that a stoichiometric engine with a three-way catalyst can provide an effective method for reducing methane emissions.

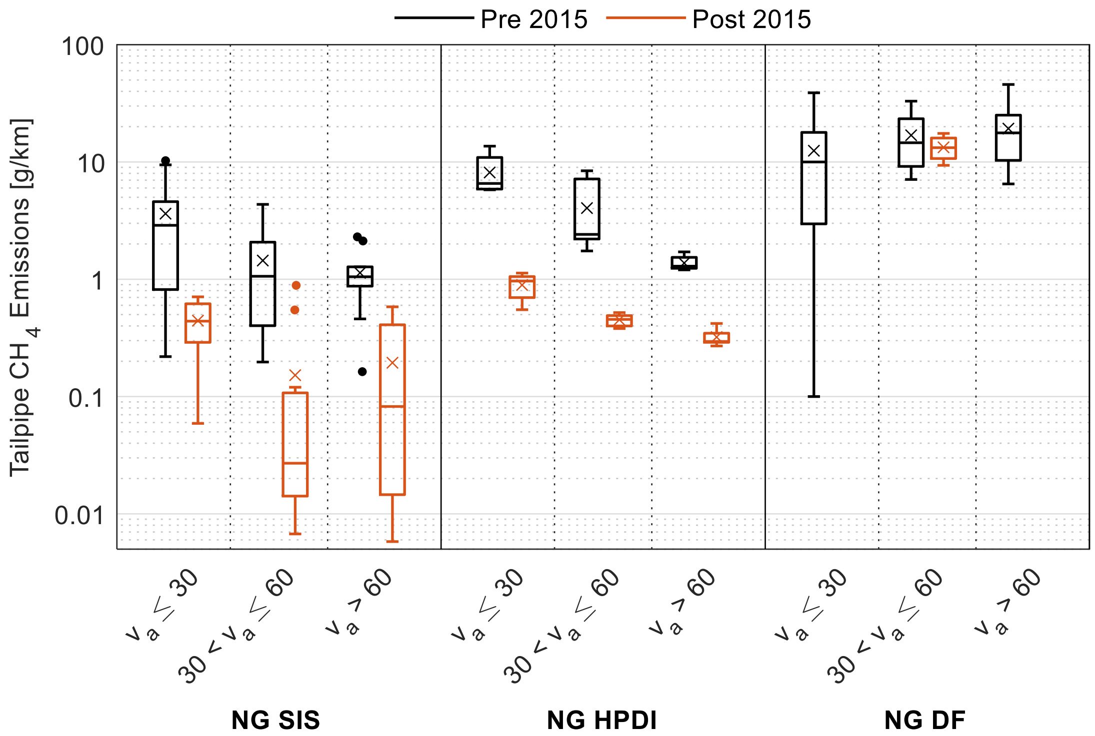

Figure 4: The distribution of tailpipe methane emissions for various natural gas engine types pre- and post-2015 for different average speeds.

Table 8: Summary of tailpipe methane emissions (quantified as methane slip) from various diesel and natural gas engines.

Model Year	Fuel Type	Engine Type	Methane Emissions [g/km]												Source
			$v_{avg} \leq 30$				$30 < v_{avg} \leq 60$				$v_{avg} > 60$				
			Mean	**Med.**	**Q1**	**Q3**	**Mean**	**Med.**	**Q1**	**Q3**	**Mean**	**Med.**	**Q1**	**Q3**	
Pre 2015 / Euro V	NG	SIS	3.623	2.884	1.130	3.893	1.444	1.082	0.494	2.051	1.135	1.050	0.913	1.193	[30, 78, 79]
	Dual	HPDI	8.139	6.558	5.940	8.757	4.046	2.413	2.260	5.917	1.375	1.293	1.270	1.398	[30, 76]
	Dual	DF	12.472	10.000	3.500	15.535	16.321	14.586	9.320	18.838	18.863	16.500	10.950	24.353	[39, 79, 82]
Post 2015 / Euro VI	NG	SIS	0.444	0.473	0.405	0.573	0.152	0.027	0.018	0.098	0.194	0.082	0.018	0.313	[80-83]
	Dual	HPDI	0.894	0.965	0.808	1.018	0.452	0.460	0.400	0.480	0.321	0.300	0.290	0.340	[84]

Dual	DF	13.235	13.235[1]	12.908	13.563	9.620[2]	[82]

1. Two data points so the median is equal to the mean; 2. One data point, therefore not included in Figure 4.

7.3.2 Crankcase Methane Emissions

Clark et al. [30] summarised all published findings on crankcase emissions for various SIS natural gas engines, which are shown in Table 9. To obtain these results, the vehicle was operated over the same route twice, once with crankcase emissions routed through the tailpipe sampling system, and once with the crankcase emissions vented to atmosphere. Delgado and Muncrief [86] suggested that if the exhaust gas recirculation for a SIS natural gas engine is on average 20%, then the methane crankcase emissions would be between 0.4-0.8%, which is within the range given for HGVs in Table 9. Since HPDI engines introduce fuel just prior to ignition, it is thought that natural gas is unable to penetrate the crevices between the piston and cylinder and crankcase methane emissions are thought to be negligible [86], though evidence for this is lacking.

Table 9: Summary of the crankcase methane emissions quantified as methane slip (%) and distance-based emissions factors [30].

Model Year	Fuel Type	Engine Type	Samples	Crankcase CH_4 Emissions [%]			
				Mean	Med.	Q1	Q3
Pre 2015 /Euro V	NG	SIS	18	0.673	0.617	0.549	0.770

7.3.3 Dynamic Venting of Methane (High-Pressure Direct Injection Engines)

HPDI engines have a dynamic venting system that is used during transient engine behaviour (sudden starting and stopping), which vents methane into the atmosphere. The only study on dynamic venting of methane in HPDI engines was undertaken by Clark et al. [30]. The estimated methane emissions due to dynamic venting by four HPDI tractors are shown in Table 10, from which it can be seen that while it is possible for no dynamic venting to occur, methane emissions can be greater than 2% of the fuel used. Other studies have also suggested that the methane emissions produced by dynamic venting could be within a similar range to crankcase methane leakage [86]. However, there is a lack of publicly available data on methane emissions by dynamic venting in HPDI engines.

Table 10: The fuel-specific methane emissions produced by dynamic venting in high-pressure direct injection (HPDI) natural gas vehicles [30].

Model Year	Fuel Type	Engine Type	Samples	Dynamic Venting Methane Emissions [%]			
				Mean	Med.	Q1	Q3
Pre 2015 / Euro V	Dual	HPDI	4	0.927	0.748	0.361	1.314

7.3.4 Summary of Total TTW Methane Emissions

Combining the evidence on methane emissions emanating from vehicle tailpipe, crankcase and dynamic venting, we summarise our best estimates for the total TTW methane emission in Table 11.

Table 11: Summary of the range of tank-to-wheel methane leakage (as a percentage of throughput) of different types of natural gas engines.

Engine Type		Methane Emissions [%]				
		Tailpipe		Crankcase	Venting	Total TTW[1]
		Pre	Post			
SIS	Central	0.441	0.305	0.673		1.001
	Low	0.194	0.002	0.361		0.364
	High	0.668	3.173	1.124		4.298
DF	Central	13.351	8.825			13.555
	Low	3.538	3.181			3.182
	High	29.156	12.58			29.157
HPDI	Central	5.075	0.237		0.927	1.487
	Low	4.995	0.170		0	0.171
	High	5.156	0.371		2.21	7.366

1. Due to the insufficient data for crankcase and venting to separate by age, the total TTWs in terms of engine types were estimated.

7.4 Nitrous Oxide Emissions

N_2O is a potent GHG with a GWP of 273 +/-130 over a 100-year time-horizon [87] and is produced by complex reactions occurring in combustion and emissions control catalysts. N_2O emissions depend on the fuel, combustion, and emissions control systems, and the combustion and catalyst temperatures. Table 12 summarises the estimated averages and standard deviations of N_2O produced by various diesel and natural gas HGVs with different engine types. Natural gas SIS engines were found to produce the lowest N_2O emissions, whereas HPDI engines appear to emit the highest levels of N_2O compared to SIS and diesel engines. While data on N_2O emissions for natural gas vehicles is sparse, it does indicate that diesel HGVs may produce lower N_2O emissions than HPDI engines. In 2002, Lipman et al. [88] reported that diesel and natural gas vehicles appear to emit the same order of magnitude of N_2O. This may no longer be the case now that modern heavy-duty diesel engines meeting the latest NO_x emissions standards are equipped with selective catalytic reduction, which can lead to significantly higher N_2O emissions than natural gas engines depending on the duty cycle [47]. The present state-of-the-art makes it difficult to definitively state whether natural gas HGVs emit more or less N_2O than diesel HGVs, particularly due to the lack of post-2015 data. All future studies should attempt quantify the N_2O emissions to thoroughly assess the overall GHG emissions of natural gas HGVs due to its high GWP.

Table 12: Summary of the nitrous oxide emissions produced by various diesel and natural gas engines.

Model Year	Fuel Type	Engine type	N_2O Emissions [g/km]								Source
			$v_{avg} \leq 30$				$30 < v_{avg} \leq 60$				
			Mean	Med.	Q1	Q3	Mean	Med.	Q1	Q3	
Pre 2015 / Euro V	Diesel	CI	0.022	0.022	0.020	0.023	0.034	0.034	0.033	0.036	[77]
	NG	SIS	0.003	0.000	0.000	0.007	0.004	0.005	0.000	0.006	[77, 89]
	Dual	HPDI	0.417	0.283	0.143	0.508	0.757	0.230	0.138	0.362	[77, 89]

7.5 Fuel Efficiency of Natural Gas Vehicles

The fuel efficiency of LNG engines compared to diesel is an important factor that influences whether LNG vehicles can reduce TTW emissions. The fuel consumption of a vehicle depends on a variety of factors including engine technology, powertrain efficiency, aerodynamic drag, load conditions, and rolling resistance, amongst others. The duty cycle also has a significant influence on fuel consumption, with urban cycles far more intensive than long-haul cycles. A summary of the fuel efficiency ranges of various natural gas heavy vehicles relative to their diesel counterparts is presented in Table 13, and it is evident that there is a significant variation in the fuel efficiency of natural gas heavy vehicles. The data suggests that there is a significant fuel efficiency penalty for natural gas engines compared to similar diesel engines. SIS shows the highest fuel consumption penalty among the natural gas engines. For pre 2015 vehicles it is up to 46% for high-speed driving, and for post 2015 vehicles, it is up to 43% for low-speed driving. Only post 2015 HPDI vehicles show the fuel consumption advantage up to 7%.

Table 13: Ranges of the fuel efficiency of various natural gas heavy vehicles relative to diesel.

Model year	Fuel	Engine type	Fuel Consumption Relative to Diesel Counterpart [%]												Source
			$v_{avg} \leq 30$				$30 < v_{avg} \leq 60$				$v_{avg} > 60$				
			Mean	Med.	Q1	Q3	Mean	Med.	Q1	Q3	Mean	Med.	Q1	Q3	
Pre 2015 / Euro V	NG	SIS	135	134	132	138	128	127	125	131	141	141[1]	136	146	[77, 78]
	Dual	HPDI	112	114	107	118	113	116	113	118					[77]
	Dual	DF	116	108	107	130	114	112	109	118	114	112	108	117	[39]
Post 2015 / Euro VI	NG	SIS	142	136	135	143	125	122	120	126	127	126	124	127	[83, 90]
	Dual	HPDI	113	111	108	115	94	94	93	96	102	97	95	105	[84]
	Dual	DF					106	106	106	106	109	109	107	111	[90]

1. Two data points so the median is equal to the mean

The ICCT stated that numerous cost-effective technologies are presently available that can deliver fuel consumption reductions in diesel vehicles of up 30-40% [86]. Improved engine technology is one avenue that can ‘contribute a significant share of the expected reductions’ [86]. Various studies have provided differing estimates of the potential improvements in fuel efficiency that can be achieved through developing advanced engine technologies, and Delgado and Muncrief [86] predicted that diesel engines will improve by around 3.5% from 2018-2025 and they assumed that natural gas engines will follow the same rate of improvement in fuel efficiency as diesel engines such that the efficiency penalty remains stable at 10% and 15% for CI and SI engines, respectively [86]. However, the authors acknowledged that potential efficiency improvements and uncertainties suggest that the likely range of the efficiency penalty relative to diesel is between 0-15%. Their modelling up to 2040 used a constant efficiency penalty of 10% and assumed the efficiency gap to diesel will remain during this time horizon [86], and is in line with the EPA’s [91] statement that natural gas vehicles will operate with a 5-15% fuel efficiency penalty relative to diesel from 2017-2025. These estimates are in line with the data for today’s technology and it is likely that the fuel efficiency of natural gas freight vehicles relative to diesel will be in the range of a 0-15% for the foreseeable future.

8 Synthesis

To synthesise this information, the review is distilled into the potential ranges of WTW methane emissions and the engine efficiency relative to diesel for SIS, DF and HPDI engines in Table 14 and Table 15. These values are used in Figure 5 to demonstrate the central estimate and expected range of WTW GHG emissions of natural gas freight vehicles relative to diesel vehicles.

Table 14: Well-to-wheel methane emissions as a percentage of throughput for three different engine technologies; central, low, and high estimates.

Engine Type		Methane Emissions [%]			
		WTP	PTT	TTW	Total WTW
SIS	Central	1.3	0.4	1.0	2.7
	Low	0.2	0.1	0.4	0.7
	High	10.0	18.3	4.3	32.6
DF	Central	1.3	0.4	13.6	15.3
	Low	0.2	0.1	3.2	3.5
	High	10.0	18.3	29.2	57.5
HPDI	Central	1.3	0.4	1.5	3.2
	Low	0.2	0.1	0.2	0.5
	High	10.0	18.3	7.4	35.7

Table 15: The energy efficiency of the different natural gas engine technologies relative to diesel.

Engine Type		Energy Efficiency Relative to Diesel, η	
		Pre 2015 / Euro V	Post 2015 / Euro VI
SIS	Central	0.75	0.78
	Low	0.66	0.62
	High	0.85	0.87
DF	Central	0.87	0.93
	Low	0.72	0.88
	High	1.02	0.95
HPDI	Central	0.89	0.97
	Low	0.81	0.76
	High	1.12	1.12

The WTW GHG emissions of diesel and natural gas vehicles are estimated by the following two equations,

$$\mathrm{WTW_{Diesel}}\left[\frac{\mathrm{gCO_2}}{\mathrm{km}}\right] = \mathrm{WTT_{Diesel}}\left[\frac{\mathrm{gCO_2}}{\mathrm{MJ}}\right]\cdot \overline{E}_{\mathrm{Diesel}}\left[\frac{\mathrm{MJ}}{\mathrm{km}}\right] + \overline{\mathrm{TTW}}_{\mathrm{Diesel}}\left[\frac{\mathrm{gCO_2}}{\mathrm{km}}\right], \quad (1)$$

$$\begin{aligned}\mathrm{WTW_{NG}}\left[\frac{\mathrm{gCO_2}}{\mathrm{km}}\right] &= \mathrm{WTT_{NG}}\left[\frac{\mathrm{gCO_2}}{\mathrm{MJ}}\right]\cdot \frac{\overline{E}_{\mathrm{Diesel}}}{\eta}\left[\frac{\mathrm{MJ}}{\mathrm{km}}\right] \\ &+ \frac{\mathrm{TTW_{NG}}[\mathrm{kgCO_2/MJ}]}{\mathrm{TTW_{Diesel}}[\mathrm{kgCO_2/MJ}]}\cdot\frac{1}{\eta}\cdot\overline{\mathrm{TTW}}_{\mathrm{diesel}}\left[\frac{\mathrm{gCO_2}}{\mathrm{km}}\right] \\ &+ \mathrm{WTW_{methane\_leakage}}\left[\%\right]\cdot x_{\mathrm{CH_4,NG}}\cdot\frac{\overline{E}_{\mathrm{Diesel}}}{\eta}\left[\frac{\mathrm{MJ}}{\mathrm{km}}\right]\cdot \mathrm{GWP_{CH_4}}[-]\end{aligned}, \quad (2)$$

where η is the energy efficiency ratio of natural gas vehicles relative to diesel counterparts. The WTW GHG emissions in Figure 5 are estimated by Eq (1) and (2) with the UK-specific emission factors [7] which are similar to the North American emissions factors [92][4]. Figure 5 demonstrates that natural gas HGVs are likely to have higher WTW GHG emissions compared to diesel. HPDI vehicles are closer to achieving parity compared to diesel due to their higher efficiency compared to other LNG vehicles even though they have higher methane leakage than SIS vehicles. SIS vehicles have relatively low WTW GHG emissions, due to lower TTW methane emissions, however they suffer from lower efficiency compared to DF and HPDI engines. For DF engines to provide lower WTW GHG emissions relative to diesel, reducing WTW methane emissions and improving efficiencies relative to diesel are a priority.

[4] The difference in TTW emissions between UK and North America is 3.6% for natural gas and 2.4% for diesel [93] Emission Factors for Greenhouse Gas Inventories, US EPA; 2018. Available from: https://www.epa.gov/sites/production/files/2018-03/documents/emission-factors_mar_2018_0.pdf., those for WTT are 0.65% for natural gas and 2.5% for diesel [94] Unnasch S, Pont J. FULL FUEL CYCLE ASSESSMENT WELL TO TANK ENERGY INPUTS, EMISSIONS, AND WATER IMPACTS. TIAX LLC; 2007..

For all three LNG engine types, Euro VI (post 2015) vehicles show higher efficiency compared to Euro V (pre 2015) vehicles. If the efficiency penalty relative to diesel is maintained, WTW GHG savings for HPDI and DF ($\eta \approx 0.9$) are likely to be in the order of 5-10%, even if WTW methane emissions are reduced to zero. If the efficiency penalty can be eliminated, a WTW methane leakage rate of above ~2.5% would negate the benefits of natural gas vehicles and lead to higher overall GHG emissions, highlighting the need to rigorously control methane emissions across the supply chain. In the best-case scenario, if the efficiency penalty is reduced to zero and methane emissions are eliminated, the full potential of the lower carbon intensity of methane would be exploited and a WTW GHG emissions reduction of ~16% could be achieved.

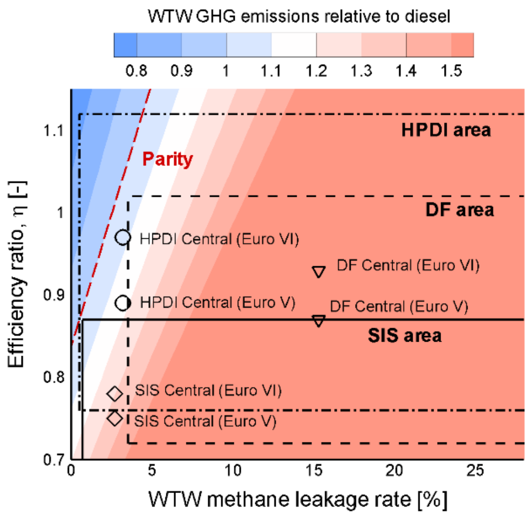

Figure 5: Total well-to-wheel greenhouse gas emissions of spark-ignited (SIS), dual-fuel (DF), and high-pressure direct injection (HPDI) natural gas heavy goods vehicle engine technologies relative to diesel and as a function of WTW methane leakage rates and η, the natural gas to diesel engine efficiency ratios.

Figure 6 compares the WTW GHG emissions for natural gas and biomethane estimated by the UK-specific data [7]. The natural gas region is equivalent to the contour plot in Figure 5 but was replotted in terms of WTW GHG emissions relative to diesel and WTW methane leakage rate for η = 0.7, 0.8, 0.9 and 1.0. The CO_2 emission factors for biomethane are defined as "net carbon zero" or "carbon neutral" according to the convention required by international GHG inventory guidelines and formal accounting rules [7], which assumes that any CO_2 emitted during the burning of the fuel (TTW) is counterbalanced by the CO_2 absorbed by the feedstock used to produce the fuel during growth. However, WTT emissions are not necessarily zero due to emissions associated with producing, processing, refining and transporting biomethane. The counterbalanced CO_2 emission is 55.28 gCO_2/MJ which is 2.4% less than NG emission factor. Due to the assumption of zero TTW emission, biomethane does lead to lower WTW GHG emissions relative to diesel and to fossil NG. While this study only focuses on biomethane to compare it with fossil NG using the same baseline data, the zero

TTW assumption also applies to the other biofuels. This indicates that the use of biofuels can potentially lead to dramatic reduction of overall WTW GHG emissions as compared to the fossil fuels. Using our best estimates of WTW methane leakages and fuel efficiency penalties, the use of biomethane would reduce WTW GHG emissions by 34%, 64%, 66% for DF, HPDI and SIS, respectively. This indicates the potential of biomethane to be beneficial in terms of overall GHG emissions despite the efficiency penalties of current engine technology. However, as indicated in Figure 9, there would be no benefit from biomethane in terms of WTW emissions if WTW methane emissions are greater than 8%. It is therefore vitally important that sources of fugitive methane emissions are carefully controlled throughout the fuel production and supply chain. Furthermore, development of infrastructure for mass production and improvement of fuel impurities are prerequisite for a wider usage of biofuels.

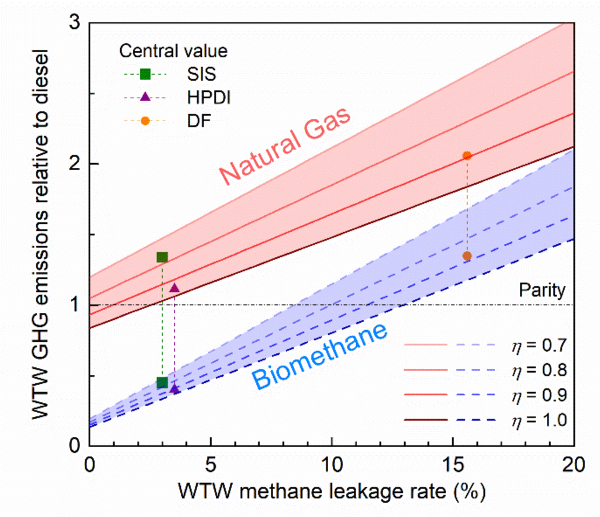

Figure 6: Total well-to-wheel greenhouse gas emissions of natural gas (NG) and biomethane relative to diesel and as a function of WTW methane leakage rates

Before 2020, higher upfront vehicle costs for NG fuelled HGVs can be compensated for by the lower fuel price relative to diesel. Langshaw et al. [95] reported that the use of LNG is financially beneficial only when refuelling at public stations; investments in private refuelling infrastructure typically negate the economic benefits. For both diesel and NG HGVs, operational expenditure including insurance, operation and maintenance costs, tolls, driver wages, and fuel costs predominates over the vehicle costs [96]. In this regard, the total cost of ownership for NG HGVs is lower than the diesel counterparts. For buses, Dyr et al. [97] suggested that the use of CNG is beneficial when the price of 1 m^3 CNG does not exceed 55% of the price of 1 dm^3 diesel fuel. However, these previous studies did not reflect the current market volatility due to the COVID and Russia-Ukraine conflict. European gas prices are now about ten times higher than their average level over the past decade [98]. Several studies present that the battery-electric trucks offer the greatest environmental and economic benefits [96, 99]. Despite high lifecycle costs and insufficient feedstock capacity, hydrogen can be an attractive alternate for regional trucks [99].

9 Conclusions and Outlook

This study provides a comprehensive review of the WTW lifecycle emissions of LNG for HGVs in comparison to diesel and is particularly focused on systematically reviewing literature on TTW emissions. TTW in-use emissions data, categorised by engine technology, model year (pre- and post-2015), and average speed of the duty cycle, is quantitatively synthesised. The two main parameters affecting the overall WTW GHG emission of natural gas HGVs compared to diesel are the fuel efficiency relative to diesel and methane leakages across the supply chain.

For WTP methane emissions, significant uncertainty in the data makes it difficult to generalise across different supply chains and geographic regions. Methane leakage rates are estimated to be 1.3% nominally, with a range of 0.2% to 10%. These WTP emissions are dominated by methane leakage during gas production (25-57%), transmission and storage (19-32%), and distribution (10-32%). For PTT, the main source of methane emissions is the venting of methane to the atmosphere due to BOG pressure build-up in the station and on-board storage tanks. The management of BOG from on-board LNG tanks, the flexibility of stations to refuel vehicles with different fuel supply systems, and the minimisation of BOG generation in the station storage tanks are critical to reducing fugitive methane emissions. Methane leakage rates for PTT are estimated to be 0.4% nominally, with a range of 0.1% to 18.3%. There is substantial variation in available data in the literature on TTW methane emissions. Several factors can explain this variation, including differences in vehicle age, catalyst temperature, engine speed, vehicle load, and duty/drive cycle. Furthermore, few measurements exist to quantify non-tailpipe TTW emissions including crankcase emissions and dynamic venting. While natural gas HGVs do typically emit less CO_2 than comparable diesel engines, the potential CO_2 benefit of natural gas is not fully exploited due to lower energy efficiencies of natural gas engines. Furthermore, methane emissions contribute significantly to total GHG emissions.

There are three primary natural gas engine types used for heavy-duty vehicles; SIS, HPDI, and DF engines. In general terms, SIS engines suffer from greater efficiency losses relative to diesel, however, tailpipe methane emissions are effectively controlled by three-way catalysts. In comparison, DF and HPDI engines have higher TTW methane emission but benefit from higher efficiencies due to compression ignition. Non-tailpipe methane emissions occur via the crankcase for SIS and DF engines, and through dynamic venting in the case of HPDI engines. The evidence regarding N_2O emissions for the different natural gas engines relative to diesel is inconclusive and requires further research. While there is evidence that air pollutant emissions from natural gas engines (particularly SIS) are lower than from diesel engines, the advancements in diesel emissions control required by more stringent emissions regulations (e.g. Euro VI in Europe) means that the air pollutant benefits of natural gas engines that did exist have been diminished.

The long-term view of the efficiency of natural gas engines relative to diesel suggests that the energy efficiency penalty will remain in the range of 0-15%, with a likely value of 10% up to 2040 without further interventions or regulatory changes [86, 91]. With this efficiency penalty, the magnitude of GHG emissions savings possible in natural gas fuelled trucks is up to 10%, which appears insufficient in the longer term when compared to climate change goals that seek to reach net zero emissions by 2050. The projection of WTW GHG emission of biomethane relative to diesel demonstrates reductions of 34%, 64%, 66% for DF, HPDI and SIS, respectively. However, controlling fugitive methane emissions in the fuel production and supply chain remains critical. Electrification is likely to lead to greater WTW GHG emissions reductions in the 2030 timeframe than are possible with natural gas HGVs [100]. Moultak et al. [100] have estimated that emissions savings of up to 60% (relative to today's diesel vehicles) could be achieved through the use of hydrogen fuel cell, electric overhead catenary charging, and electric induction charging based vehicles in China, Europe, and the US in 2030.

Given the efficiency penalty and methane leakage, natural gas offers no significant benefits over diesel, which is in line with the implication from many other studies [22, 35, 43-45, 95]. Future research is needed to better identify the levels of methane emissions throughout the WTP process such as natural gas recovery and pipeline transport. Also, the impacts of non-tailpipe TTW emissions such as crankcase emissions and dynamic venting from LNG vehicles need further investigation. To resolve the uncertainty in the pollutant emissions associated with natural gas HGVs such as N_2O emission, more evaluation of the air quality benefits or drawbacks is required for better understanding of the trade-offs associated with potential alternatives for future HGVs.

List of abbreviations

BOG	Boil-off gas
CI	Compression ignition
CNG	Compressed natural gas
CO	Carbon monoxide
DF	Dual fuel
EU	European Union
GHG	Greenhouse gas

GWP	Global warming potential
HDV	Heavy duty vehicle
HGV	Heavy goods vehicle
HPDI	High-pressure direct injection
LNG	Liquefied natural gas
NG	Natural gas
NOx	Nitrogen oxides
PBU	Pressure build-up unit
PM	Particulate matter
PTT	Pump-to-Tank
SI	Spark ignition
SIS	Spark ignition stoichiometric
TTW	Tank-to-Wheel
WTP	Well-to-Pump
WTW	Well-to-Wheel

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