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Published in: Proceedings of the 30th CIMAC World Congress 2023

Published: 12/06/2023

Document Version Publisher's final version

License Other

Link to publication

Please cite the original version: Kuittinen, N., Heikkilä, M., Jalkanen, J-P., Aakko-Saksa, P., & Lehtoranta, K. (2023). Methane slip emissions from LNG vessels - review. In *Proceedings of the 30th CIMAC World Congress 2023* Article 629



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Methane slip emissions from LNG vessels - review

Emission Reduction Technologies - Exhaust Gas Aftertreatment Solutions

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This paper has been presented and published at the 30th CIMAC World Congress 2023 in Busan, Korea. The CIMAC Congress is held every three years, each time in a different member country. The Congress program centres around the presentation of Technical Papers on engine research and development, application engineering on the original equipment side and engine operation and maintenance on the end-user side. The themes of the 2023 event included Digitalization & Connectivity for different applications, System Integration & Hybridization, Electrification & Fuel Cells Development, Emission Reduction Technologies, Conventional and New Fuels, Dual Fuel Engines, Lubricants, Product Development of Gas and Diesel Engines, Components & Tribology, Turbochargers, Controls & Automation, Engine Thermondynamis, Simulation Technologies as well as Basic Research & Advanced Engineering. The copyright of this paper is with CIMAC. For further information please visit https://www.cimac.com.

ABSTRACT

The International Maritime Organization (IMO) regulations on emissions of nitrogen oxides (NOx) and sulphur oxides (SOx) have lead to increased utilization of liquefied natural gas (LNG) as a fuel for shipping. Due to very low sulphur content in LNG, the contribution to SOx emissions is negligible. NOx emissions depend on the engine combustion cycle and with LNG engines utilizing otto-cycle (or diesel cycle engines with post combustion treatment) also the strict IMO Tier III NOx limit can be achieved. In addition, it is shown that LNG utilization leads to significantly lower particle emissions compared to liquid marine fuels. Thus, LNG utilization has direct effects and indeed benefits on air quality and human health. Moreover, CO2 emission can be reduced with LNG use compared to diesel fuels, since LNG is mainly composed of methane with a higher H/C ratio compared to diesel. The hydrocarbon emissions, on the other hand, are higher with LNG compared to diesel fuels and mostly include the main component of LNG, methane. This 'methane slip' should be minimized because methane is a strong greenhouse gas and reduces the benefit of lower CO2 emissions.

While the formation of methane slip is known to result from LNG combustion, there has been a lack of knowledge of the methane slip emission's magnitude from the LNG engines. In this review paper, methane slip values are collected from the current literature and ship owner data is utilized to complement the data with engines from recent years. This will contribute to understanding the methane slip from the current LNG fleet.

High-pressure 2-stroke slow speed (diesel cycle) engines already show very little methane slip today, while higher methane slip values are reported for low-pressure dual fuel engines. Out of 614 vessels with an identified LNG engine, the low-pressure dual fuel concept (either 4-S or 2-S) is also the most popular LNG engine technology found in 78.1% of the ships, while high-pressure dual fuel technology is found in 14.8% of the ships and lean burn spark ignited engines in 1.7%. This is reflected in the amount of methane slip data found in the scientific literature which focuses on low pressure dual-fuel engines. The engine load has a significant effect on the methane slip formation. In general, the lower loads tend to increase the methane slip formation compared to higher engine loads.

1 INTRODUCTION

The use of liquefied natural gas (LNG) as shipping fuel has increased in recent years which improves the air quality and reduces detrimental human health impacts of air emissions. According to a recent report, about 20% of the total vessel orders in 2021 were LNG-fueled [1]. Vessels using LNG as fuel enable one transition pathway from fossil to non-fossil fuels. In such a transition, methane molecules, which are the main component of LNG can be considered as a drop-in fuel, compatible with existing marine engines. Methane may be of fossil, bio- or synthetic origin, but non-fossil methane needs to be produced in large enough quantities. Further, the origin of the biomaterial and electricity used in fuel synthesis must be sustainable. LNG can be considered as a transition fuel which facilitates the decarbonization of maritime transport. Ideally, existing engine solutions and tank arrangements can be used with very low or zero carbon fuels with minimal need of modifications [1], [2]. Dual-fuel (DF) engines provide fuel flexibility for ship operators, because both liquid and gaseous fuels can be used.

LNG is mainly composed of methane (CH₄) which has higher hydrogen to carbon ratio and energy content compared to liquid fuels, leading to lower emissions of carbon dioxide (CO₂). The use of LNG as marine fuel can also reduce the emissions of nitrogen oxides and particulate matter including black carbon, relative to operation on marine gas oil [3]–[6]. In comparison to heavy fuel oil, natural gas combustion has been estimated to produce a unit of energy with 24% less carbon dioxide emissions, 90-99% less sulfuric oxides, and 90% less particulate matter [7]. While the use of LNG has benefits in terms of CO₂ emissions and local air pollutants, emission of unburnt methane to atmosphere remains a concern.

In the atmosphere, methane is an important greenhouse gas (GHG) contributing to climate change. On instantaneous basis, methane is 120 times stronger absorber of infrared radiation than CO₂, but its atmospheric lifetime of 12 years is shorter compared to CO₂ which can remain in the atmosphere hundreds of years. Considering a 100year timescale, methane has 29.8 times greater global warming potential (GWP) than carbon dioxide and is 84 times more potent on a 20-year timescale [8]. Thus, methane emissions are considered highly relevant to 2050 climate objectives [9]. In addition to being a greenhouse gas, methane contributes to tropospheric ozone formation which is an air pollutant. Due to the high GWP of methane, even low emissions may negate the CO₂ benefits of switching to LNG from heavy fuel oil or marine diesel oil. [10].

Methane emissions occurring from the carriage and consumption of LNG as fuel in marine vessels can occur as fugitive emissions, through venting, or via methane slip from the engines. In a recent study on-board an LNG carrier during a transatlantic voyage, it was found that methane slip from main and auxiliary engines accounted 99% of methane emissions across the voyage and 35% of total GHG emissions of the voyage on this specific ship (Tankto-Wake, TtW), representing a significant opportunity for mitigation through the study and development of engine technology. In a study considering the total Well-to-Wake (WtW) GHG emissions of LNG fueled engines by applying extensive life cycle analysis approach and data from several engine manufacturers, unburned methane was reported to contribute up to 22% of the WtW GHG emissions, methane slip during combustion (TtW) accounting for 16%, and methane emissions in the supply chain (Well-to-Tank, WtT) 6% of the total GHG emissions [11]. A need for reducing TtW methane slip emissions has also been recognized to increase the viability of electro- and biomethane fuel pathways [7]

Globally, LNG use as maritime fuel is regulated within the International Convention for the Prevention of Pollution from Ships and the International Code of Safety for Ships Using Gases or Other Low-flashpoint Fuels (IGF Code), which defines the needed technical requirements and procedures for carriage and combustion of natural gas as marine fuel. Neither regulation limits methane emissions as product of combustion, nor are methane emissions counted in any regulations set by the International Maritime Organization concerning GHG emissions or energy efficiency. However, the IGF Code stipulates that venting fuel vapor from LNG tanks is not allowed except in an emergency. The transformation of parts of liquefied fuel into gaseous form (so called boil-off) by the ambient temperature should be controlled by either reliquefying, combustion, pressure accumulation, or by cooling of the liquefied gas fuel [12].

Currently, the European Union (EU) is in the process of issuing amendments for directives that regulate maritime transport as part of the Green Deal and Fitfor55 environmental packages. Two specific regulations will affect methane emissions from ships: the Emissions Trading System (ETS) and the FuelEU Maritime. Both have passed the EU Parliament vote and are currently being negotiated between the European Commission, Parliament, and member states. Both mechanisms will include carbon dioxide, methane, and nitrous oxide (N₂O) GHG emissions from the maritime transport [13], [14]. ETS will consider emissions on TtW basis and the FuelEU Maritime, the effect of methane slip

is introduced as mass percentage of the fuel used by the engine [7]. Additionally, global attempt to mitigate methane emissions was agreed on in the COP26 United Nations Climate Change Conference where the United States and the EU together with 100 other countries signed their Global Methane Pledge, aiming to cut methane emissions by 30% by 2030 compared to 2020 levels [15].

This paper focuses on collecting information on the methane emissions that originate when unburned methane exits the ship engines (methane slip). Methane slip reduction has been recognized as a topic for engine manufacturers for over 10 years and mitigation of methane slip is enabled both by developing engine technology, after-treatment technologies as well as vessel specific systemic solutions [7]. However, numerical data of methane slip from marine engines in the literature is scarce, especially for the engines build in the recent years, posing a challenge for the estimation of total methane slip emissions from shipping today and constructing scenarios for the future. The aim of this work is to provide an overview of the published methane slip emission factors (EFs) as well as to complement the existing values by collecting ship owner data covering measurements conducted on newly build engines (years 2019-2022).

2 METHODS

The modular Ship Traffic Emission Assessment Model (STEAM) [16] developed at the Finnish Meteorological Institute (FMI) was used to identify the number of LNG powered vessels currently in operation and the shares of different LNG engine types installed. STEAM combines global Automatic Identification System (AIS) data both from satellite and terrestrial sources (obtained from Orbcomm Ltd.) with a database that is updated using the IHS Markit ship information service. Among other data, the database contains information on each vessel's main and auxiliary engine fuel type and engine model. The engine model is then connected with a database containing information of known LNG engine models. Currently, the LNG-engine model database contains 323 engine models with their respective model code, engine manufacturer, Break Specific Energy Consumption (BSEC) at 80% engine load, engine speed in rpm, LNG engine type, and the engine load below which engine operates only on liquid fuel. STEAM modelling results and AIS data from 2021 were used for this review.

Information of LNG use as maritime fuel was collected from the International Maritime Organization Data Collection System for fuel oil consumption on ships (DCS), which has been storing reports since 1st of January 2019 [17]. At the time of the study, reports for years 2019, 2020, and 2021 were available. As per the reports, 27 221 vessels out of 32 511 (84%) under the scope reported their fuel consumption to DCS in 2019, 27 723 vessels out of 32 558 in 2020 (85.1%) and 28 171 out of 32 998 (85.4%) in 2021. Therefore, the absolute reported fuel masses do not represent 100% of fuel combusted at sea. LNG-powered vessels sailing in Europe were investigated using the EU Monitoring, Reporting and Verifying (MRV) open-access database (mrv.emsa.europe.eu). Reports from 2018-2021 were available, and the EMSA dataset version 90 was used for 2021.

Information of methane slip as well as other exhaust emissions from LNG engines were collected from published literature as well as by including ship owner data for engines from recent years. Emission factors for methane were collected in brake specific terms (g/kWh). The engine construction years for which methane slip results have been reported in the literature are 2010 (and older / newer), 2012, 2013 (and newer), 2016, 2016 (retrofitted), and 2021. One of the main aims of this paper is to complement the reported methane slip data with values from engines designed and built in the recent years (2019-2022) to provide an understanding of the current methane slip status engines. The from modern LNG engine construction year is taken directly when reported, and in other cases the building year of the vessel is assumed. If neither engine construction year nor building year of the vessel is reported, publication year is indicated (marked as publ.). It should be noted that the engine construction year does not always indicate the technology level, because old engine technology can be installed in new ships.

In this paper, the LNG engines available in the marine market are divided in four categories according to the engine type:

- Type 1: Lean Burn Spark Ignited engines (LBSI)
- Type 2: 4-stroke Low Pressure Dual Fuel engines (LPDF 4-S)
- Type 3: 2-stroke Low Pressure Dual Fuel engines (LPDF 2-S)
- Type 4: 2-stroke High Pressure Dual Fuel engines (HPDF 2-S)

Type 1 engines include Lean Burn Spark Ignited (LBSI) engines utilizing only natural gas as fuel with spark plug ignition. The LBSI engines are typically 4-stroke high or medium speed engines utilized in smaller vessel types and where fast engine response is needed [11]. Type 2 category includes 4-stroke Low Pressure Dual Fuel (LPDF 4-S) engines where natural gas is injected in low

pressure during the compression stroke of the engine and small amount of liquid fuel is used for ignition. The LPDF 4-S engines typically operate with medium speed and typically allow more flexible operation than their 2-stroke counterparts. They are mostly utilized in ferries, cruise ships, and short sea shipping as well as auxiliary engines at large vessels [11]. Low Pressure Dual Fuel engines operating with 2-stroke cycle (LPDF 2-S) are categorized as Type 3. The LPDF engines operate according to the thermodynamic Diesel cycle in liquid fuel mode and according to Otto cycle in LNG mode, therefore in some sources they are also referred to as Otto-DF engines. Finally, Type 4 engines include 2-stroke High Pressure Dual Fuel engines where natural gas is injected in high pressure at the end of the compression stroke, simultaneously with the liquid pilot fuel injection. These engines are categorized as slow speed and operate according to Diesel cycle also in dual-fuel mode therefore they are also referred to as Diesel-DF engines. The 2-stroke engine types have the highest efficiency and power and are commonly used in large ocean-going cargo ships [11].

LPDF 4-S engines were found to be most widely covered in the scientific literature with at least one publication including results for LPDF 2-S engines. As seen later in Section 3.3, LPDF 4-S and LPDF 2-S engines are most widely used and therefore these engine types were also in the main focus of the review. LBSI engines are mainly used in smaller vessel categories, but their methane slip was included when reported in the publications. Original measurement data for HPDF engines were found to be lacking from the scientific literature but few values given by engine manufacturer were included in reports. Generally, the methane slip from HPDF engines is considered low due to the applied injection and combustion method.

The choice of engines for a vessel is dependent at least on the ship type, size, and operational parameters. Large container ships often use 2stroke engines and mainly operate in deep-sea regions where they may apply constant engine load for long periods after exiting the harbor. On the other hand, ferries or cruise ships are typically equipped with 4-stroke engine and operate on coastal areas where engine load changes may be more frequent. [11]. To consider these varying operational patterns with different engine types, the methane slip values are presented as function of engine load percentage when the load-related data is available.

3 LNG SHIPS AND ENGINES

3.1 LNG as marine fuel

Natural gas consists primarily of methane, but its composition varies according to region with varying share of alkanes such as ethane, propane, and isobutane. For transport overseas and use as marine fuel, natural gas is liquefied in a process that cools the pre-treated gas down to a liquid form in -162°C. Liquefaction allows storing natural gas in 600 times lower volume than gas in standard atmospheric pressure. [18], [19]. Ushakov et al. [20] collected the composition of LNG from several suppliers in Europe, where the methane content varied between 91-96%, ethane content between 3-7%, and propane content between 0.3-1.4%. Methane content in LNG sources from the different exporters can vary from 87.3 to up to 99.7%, ethane between 0.09-9.97%, and propane between 0.03-3.3%. [21]

Table 1. LNG use as maritime fuel and the share of LNG use by vessel type as reported to IMO in 2021.

Vessel type	LNG use (t)	Share (%)
Bulk carrier	36 773	0.3
Combination carrier	0	0.0
Container ship	163 707	1.3
Cruise ship	59 796	0.5
Gas carrier	2 137 002	16.9
General cargo ship	4 052	0.0
LNG carrier	9 958 661	78.9
Others	40 203	0.3
Passenger ship	3 551	0.0
Refrigerated cargo	0	0.0
Ro-Ro cargo ship	6 166	0.0
Ro-Ro cargo ship	16 895	0.1
Ro-Ro passenger ship	94 802	0.8
Tanker	101 513	0.8
All vessel types	12 623 121	

According to the IMO DCS data, total reported use of LNG as maritime fuel was 10.48 Mt (metric) in 2019, 11.97 Mt in 2020 and 12.62 Mt in 2021 [17]. Out of all combusted maritime fuel, this represents a share of 4.9% in 2019 (total 213.03 Mt), 5.9% in 2020 (total 203.10 Mt) and 5.9% in 2021 (total 212.23 Mt). These numbers cover the ships with an IMO registry number over 5000 gross ton who went through the DCS reporting. Most of the total LNG combusted was used by LNG carriers (71.5% in 2019, 77.5% in 2020 and 78.9% in 2021) then followed by gas carriers, which are vessel carrying gaseous cargo such as propane (26.1% in 2019, 19.2% in 2020 and 16.9% in 2021). The remaining vessel types represented 2.4% of all LNG combusted in 2019, 3.3% in 2020 and 4.2% in 2021. LNG use by vessel type is presented in Table 1. LNG use increased by 14.2% in 2020 compared to 2019 and by 5.4% in 2021 compared to 2020.

LNG powered vessels sailing in Europe that reported to the MRV database were identified by having a carbon factor (CF) of less than 3.0 (Table 2). The CF was calculated by dividing the reported total annual CO₂ emissions with the reported total annual fuel consumed. Liquefied fuel oils have a CF > 3.0 and the CF of LNG is 2.7.

Vessel type	All	CF < 3.0
Bulk carrier	3714	3
Chemical tanker	1386	11
Container vessel	1825	28
Container / Ro-ro	64	0
Gas carrier	327	14
General cargo ship	1236	2
LNG carrier	287	247
Oil tanker	1847	16
Other vessel types	149	2
Passenger ship	107	5
Refrigerated cargo carrier	153	0
Ro-pax ship	369	16
Ro-ro ship	224	0
Vehicle carrier	464	3
All ships	12152	347

Table 2. Ships that reported to the MRV database in 2021 and that had a carbon factor below 3.0.

3.2 LNG vessel types

The FMI STEAM model and its databases were used to identify the number of LNG powered vessels currently in operation and the shares of different LNG engine types installed. The FMI ship database currently contains data of 113 133 vessels. Out of these, 614 vessels are equipped with a known LNG-powered DF engine and 35 vessels were marked as using LNG for propulsion with an unknown engine model.

By vessel type (Figure 1), the largest share of known dual fuel engines was installed on LNG tankers (55.2% of all LNG-powered DF engine ships) followed by production tankers (7.3%), container vessels (5.5%), ro-ro vessels (4.9%), and crude oil tankers (4.9%). LNG tankers are known to use their transported cargo as fuel for propulsion and auxiliary needs. There are 707 LNG tankers in the FMI ship database out of which 666 were transmitting AIS data in 2021. The remaining LNG

tankers (368) that did not have a known dual-fuel engine mostly have a steam turbine main engine, some have a gas turbine and a few have a conventional oil engine. This is in line with the LNGindustry's own reporting. The International Gas Union annual report (International Gas Union, 2022) states that there were 641 active LNG tankers by the end of April 2022 including 45 floating storage regasification units and 5 floating storage units. An interpretation of the IGU and other reports by American Bureau of Shipping gives a total of 694 LNG-fueled ships either in operation or under construction as well as 213 more estimated to be LNG-ready [1].



Figure 1. Share of known dual fuel engines in different vessel types from STEAM.

Wartsila LPDF 4-S engines hold the largest share of installed DF engines (287 vessels or 46.7% of all LNG-powered DF engine ships). WinGD LPDF 2-S engines were installed on 157 (25.6%) vessels. MAN engines were installed on 113 (18.4%) vessels out of which 17 had a LPDF 4-S and 96 a HPDF 2-S engine. The remaining manufacturer shares of dual fuel engines installed on vessels were: ABC (5.7% of all LPDF 4-S engines), Caterpillar (2.9%), Mitsubishi (1.7%), Niigata (1.1%), Hyundai (0.9%), MTU (0.6%) and Yanmar (0.3%). All installed Type 1 (LBSI) engines were manufactured by Bergen, all Type 3 (LPDF 2-S) were WindGD and all Type 4 (HPDF) were MAN. Out of the installed Type 2 (LPDF 4-S) engines, 82% were manufactured by Wartsila, 5.7% by ABC, 4.9% by MAN, 2.9% by Caterpillar, 1.7% by Mitsubishi, and 1.1% by Niigata.

3.3 LNG engines in operation

Out of the 614 vessels with an identified LNG engine, 11 were with Type 1 (LBSI) engine (1.7%), 350 with Type 2 (LPDF 4-S) engine (53.9%), 157 with Type 3 (LPDF 2-S) engine (24.2%) and 96 with Type 4 (HPDF) engine (14.8%). The distribution of engines by type is shown in Figure 2.

Out of the 614 dual fuel engine vessels in the FMI database, 508 were transmitting AIS data in 2021 and 588 vessels were modelled with STEAM as

using LNG for propulsion. Maximum installed main engine power on the LNG-powered vessels were less than 5 000 kW (13.0% of the vessels), between 5 000 and 10 000 kW (14.3%), between 10 000 and 25 000 kW (25.4%), between 25 000 and 50 000 kW (41.2%) and more than 50 000 kW (6.0%), showing majority of ships with LNG engine to have installed main engine power between 25-50 MW.



Figure 2. Distribution of engines by type in vessels identified with an LNG engine from STEAM.

Measured by the total installed main engine power (Figure 3), engine sizes below 5 MW represent 2% (installed power 298 MW), 5-10 MW engines 4% (629 MW), 10-25 MW engines 20% (3 026 MW), 25-50 MW engines 59% (8 990 MW), and above 50 MW engines 15% of the total (15 246 MW).



Figure 3. Distribution of total installed main engine power in LNG powered vessels by main engine power class.

3.4 Methane slip mechanisms

In LBSI engines (Type 1), the air-fuel mixture is ignited by spark plugs whereas in LPDF engines (Types 2 & 3) the ignition occurs as liquid diesel fuel (pilot fuel) is injected into the cylinder. In both engine types, the occurrence of methane slip can be explained either by temporary hiding of methane in cylinder crevices or quenching, both of which lead to a fraction of the injected natural gas to exit the engine unburned.

As natural gas is injected in low pressure during the early compression stroke and has time to reside in the cylinder before ignition, it may be pushed to the crevice volumes of the cylinder, including e.g., the gasket area between the cylinder head and cylinder liner, and remain unignited until slipping out of the cylinder during exhaust stroke. The mixing of the injected gas with air may also happen unevenly, creating gas-rich and gas-lean regions in the cylinder where also varying temperatures are reached.

Quenching can be explained as methane locally cooling down rapidly in the coldest areas of the combustion chamber, and as it requires a high temperature of >600°C to autoignite, remaining unburned. [7], [22]. The lean combustion and consequent low thermal load may make the engine susceptible to quenching, especially at low engine loads when fuel-air ratio is low ([23]-[25], according to [2]). Sommer et al. [26] showed an increase of methane emissions as a function of the air-fuel ratio, as operation at low loads results in leaner airfuel mixtures and attributed the methane slip to decreased flame speeds in the lean mixtures. Methane slip may also be caused in LBSI and LPDF engines by direct slip due to valve overlap. If exhaust valve is partly open during gas admission, fraction of methane may flow directly to exhaust stack. In LPDF engines applying prechambers, incomplete combustion in the prechamber may be one additional source of methane slip [27].

More factors mentioned to affect methane slip are changes in natural gas composition and ambient conditions since fuel composition and temperature both affect the optimal air-to-fuel ratio. During fluctuating load conditions (such as heavy weather) the turbocharger may also fail to follow changing air demand which may also alter methane slip. Early or incomplete combustion leading to altered methane slip may also occur due to pre-ignition of lubrication oil or dripping fuel atomizers. [7]. In addition, evaporation from the lubricant oil film is stated as a known source of total hydrocarbon emissions which results from desorption of any formerly absorbed fuel molecules [27].

In HPDF engines, natural gas is injected in high pressure together with the pilot fuel and combustion temperature is higher, therefore leading to lower susceptibility to quenching. Also, as the methane burns as it is being injected, there is less opportunities for methane residing in the crevice volumes. One potential phenomenon that could affect methane slip in HPDF engines is local flame extinction due to high turbulence. The low methane slip from HPDF engines can be seen as trade-off with NO_x emissions since the higher combustion temperature leads to NO_x levels requiring after-treatment to be IMO Tier III compliant, while LPDF engines do not require any after-treatment in gas mode. [22], [27].

Recognized engine technologies for reducing methane slip include engine component design (e.g. minimizing crevices, piston ring design), high pressure injection (requiring gas compression equipment), engine tuning and control software (optimizing: valve timing, combustion timing, cylinder cut-off to enhance combustion velocity and decrease quenching, pilot fuel injection timing and quantity) as well as exhaust gas recirculation [7]. The engine technologies can be applied either to new-build engines or as an upgrade to existing engines, but methane abatement has also been studied by utilizing after-treatment systems such as plasma reduction systems (application of highvoltage current to convert methane to CO and H_2O) or methane oxidation catalysts (requiring means to avoid sulfur poisoning of the catalyst). In addition, vessel specific system-based solutions, such as energy efficiency technologies, batteries, and shaft generators have been suggested to reduce total methane emissions [7].

4 EMISSIONS FROM LNG ENGINES

4.1 Reported data

The amount of methane slip data from LNG engines is limited and relies on measurement data in test-bed [5], [20] or data provided by engine manufacturers [2], [7], [11], [19], [25], [28] although a handful of scientific studies [4], [6], [10], [20], [26], [29], [30] which report original research data collected during on-board measurements are recognized. For the purposes of this study, ship owner data from 6 LPDF 2-S and 5 LPDF 4-S engines from years 2019-2022 were received to complement the data with methane slip information from recently build engines.

In the publications which reported the applied measurement method for methane, (heated) flame ionization detection ((H)FID) analyzer was most typically used [4], [6], [20], [26], [29], in one case combined with gas chromatography [4]. Sommer et al. [26] noted that since FID is sensitive to all hydrocarbons, the efficiency of the catalytic cutter used to isolate other hydrocarbons from methane must be considered when reporting results. Fourier transform infrared spectroscopy has also been used in combination with gas chromatography [5], [30] as well as stand-alone device in continuous emission monitoring system (CEMS) [10]. Sommer et al. [26] also introduced the use of custom-built methane sensor utilizing wavelength modulation spectroscopy (WMS). In cases where data originates from non-peer-reviewed scientific literature such as from engine manufacture or ship owner, specific details of the measurement method are not available. It is recognized that more uniform guidelines for measuring methane concentration in

the exhaust, converting it to emission factors as well as reporting could be beneficial.

If methane emissions are not measured directly, they may be reported as part of total or unburned hydrocarbons. Methane has been experienced to comprise 80% to 95% of unburned hydrocarbon emissions according to engine manufacturers, however this is highly dependent on the gas quality [11]. In the on-board study by Anderson et al. [4] around 85% of the total hydrocarbons were found to be methane whereas Ushakov et al. [20] reported 92-97% in their study. Lehtoranta et al. [31] reported that at 85%/40% loads the total hydrocarbons (THC) in the exhaust contained 96.7%/96.5% of methane. 2.2%/2.3% ethane and 0.29%/0.13% propane, very close to the composition of the natural gas used in the study (96.4% methane, 2.3% ethane, 0.35% propane).

The methane slip from LNG engines can be described in various units. Firstly, ratio of fuel loss compared to total fuel consumed by the engine can be expressed as percentage, in which case LNG composition should be known to determine the amount of slipped methane. Pavlenko et al. [19] also mention reporting of methane slip as mass of methane per volume of LNG or per the energy content of the available fuel. In this report, the methane slip is expressed in brake specific basis, as mass of methane per useful shaft work of the engine (g/kWh), enclosing the efficiency of the engine. This was also the most common method of reporting found in the included literature. In the cases where methane emissions were expressed as CO₂ equivalents (gCO₂eq/kWh), the 100-year CO₂ equivalent of 36 applied in the specific study [10] was used for conversion.

4.2 Methane emission factors

4.2.1 Methane slip of different sized engines

For LPDF 4-S and 2-S engines, emission factors as function of engine power are shown in Figure 4 and Figure 5. For LBSI engines, 7 engines were included in the study by Ushakov et al. [20] but engine size (1.46 MW) was included only for one of the engines, for which EFs ranged from 3.7-27.6 g/kWh. For HPDF engines no values related with specified engine size were found.

For LPDF 4-S engines, EFs were reported for engine sizes between 1.4-7.6 MW. Majority of reported values were between 0-30 g/kWh, but also increased values up to 269 g/kWh were observed, attributed to low load conditions as seen later in Figures 6-7. Majority of the engines had maximum rated power below 4.4 MW with only one engine sized at 7.6 MW. While at each load, the methane slip seen for the 7.6 MW engine is among the lowest, due to the single data point in this range and large variation between 3-4.4 MW engines, definite conclusions of the effect of engine size are not possible.



Figure 4. Methane EFs reported for LPDF 4-S engines with varying engine sizes at 23-30%, 40-50%, and 70-75% load points. Dark tones are used for engines from years 2020-2023, midtones for engines from years 2016-2019 and light tones for engines from years 2015 or older. Measurements conducted in testbed are marked with asterisk. In cases, when exact engine size is not known (triangles), range (e.g. <4MW) is given in the label but result is plotted at the upper limit (e.g. 4MW).

It should be noted that for part of the engines, only upper limit of the engine size range is known, and thus smaller engine size may be one explanation to the variation in EFs seen at 23-30% load and 40-50% load. For engines below 4.4 MW, with one exception, the new engines from 2020-22 give the lowest emissions together with an engine piloting new combustion concept [30].





For LPDF 2-S engines, EFs were found for engine sizes of 11.53 MW, <12 MW and >50 MW. Methane

slip emissions were mainly between 2.4-3.6 g/kWh, but emissions at low load point as seen later in Figures 6-8 could range up to 7.2 g/kWh. Generally, the load dependent variation in methane slip EFs seems to be suppressed in larger engines, where each load condition gave EFs between 2.5-3.1 g/kWh. Overall, the variation among methane slip at certain load and engine size is smaller for LPDF 2-S engines, possibly because these engines are from recent years and probably from a single manufacturer. In comparison to LPDF 4-S engines, LPDF 2-S produce lower methane slip, especially at low loads.

4.2.2 Methane slip as function of engine load

Methane slip emission factors as a function of engine load were most often reported as brake specific emission factors (g/kWh). Figure 6 shows the methane slip values for 7 LBSI 4-S engines, 16 LPDF 4-S engines, and 7 LPDF 2-S engines.



Figure 6. Methane emission factors as a function of engine load for all engine types. Green color is used for LPDF 4-S engines, purple for LPDF 2-S engines and yellow for LBSI 4-S engines. Measurements conducted in testbed are marked with asterisk. If engine year is not available, publication year (indicated by 'publ.') is used instead. Note logarithmic scale.

For LBSI engines, EFs were found for engine loads between 10 and 100% whereas in the case of

LPDF 4-S engines, values were found for 0-100% loads and in the case of LPDF 2-S engines for 25-100% loads. For HPDF engines, no load specific values were found. However, a recent report gave methane EF ranges of 2.4-5.8 g/kWh for LPDF 4-S engines with engine load above 50%, and 1.6-2.3 g/kWh for LPDF 2-S engines without EGR, 1.1-1.6 g/kWh for LPDF 2-S with EGR, and 0.2-0.28 g/kWh for HPDF 2-S engines for engine load ranges between 25-85% [7].

Increase in brake specific methane emissions as function of decreasing engine load can be observed with all engine types. For LBSI 4-S engines at 100% load, EFs range between 2.5-4.2 g/kWh when the range at 75% load is 3.3-5 g/kWh, at 50% load 4.1-7.2 g/kWh and at 10% load 6.4-42 g/kWh. Respectively, for LPDF 4-S engines (shown separately in Figure 7), the ranges are 1.5-10.1 g/kWh at 100% load, 3.1-10.1 g/kWh at 75% load, 2.6-16.2 g/kWh at 50% load, 6.1-70.2 at 25% load and 12.2-109 g/kWh at 10% load. For an engine piloting new combustion concept [30], emission factors of 1.4-3.9 g/kWh were reported at 100%-10% loads. Again, in the case of LPDF 2-S engines (Figure 8), reported EFs for between 1.9-2.5 g/kWh for operation at 100% load, 2.4-2.9 g/kWh at 75% load, 2.4-5.1 g/kWh at 50% load and 2.8-7.2 g/kWh at 25% load.



Figure 7. Methane emission factors as a function of engine load for LPDF 4-S engines. Dark tones are used for engines from years 2020-2023, midtones for engines from years 2016-2019 and light tones for engines from years 2015 or older. Measurements conducted in testbed are marked with asterisk. If engine year is not available, publication year (indicated by 'publ.'). Note logarithmic scale. The engines were classified to three categories representing the newest engines from years 2020-2023, engines from years 2015-2019 and engines from year 2015 or older (indicated with different tones in figures above). For the newest engines, data originated either from ship owner or the recent studies by Balcombe et al. [10] and Lehtoranta et al. [30] whereas for the engines before 2015, data originated mainly from the study by Anderson et al. [4] and the data set presented by Stenersen & Thonstad [25] and Ushakov et al. [20]. The engines presented in the latter set of data as post-2013 were labeled to be in the 2015 or older category since the exact year is not known, however they could be from 2016 or 2017, the year of the first publication.



Figure 8. Methane emission factors as a function of engine load for LPDF 2-S engines. Dark tones are used for engines from years 2020-2022 and midtones for engines from years 2016-2019. Measurements conducted in testbed are marked with asterisk. If engine year is not available, publication year (indicated by 'publ.') is used instead.

In part of the publications, methane slip values were found expressed as the weighted EFs over the ISO 8178 E3/E2 test cycle used for reporting NOx emissions. However, Lindstad et al. [2] presented that today's vessels typically operate at less than 75% power, suggesting that the use of E2/E3 cycle may underestimate the methane slip compared to real-life operation. Based on STEAM model data from 2021, the average main engine load for ships using LNG engines is 60.5%. In their study, Peng et al. [6] reported methane slip emissions weighted over the E2 cycle but also over actual ship activity profile for a vessel that was operated in harbor service, not at open sea. They noted that the actual weighting factors that should be used to depict the operation of the specific vessel were significantly different from the E2 cycle and for that reason, the EFs of methane and total hydrocarbons weighted according to the E2 cycle resulted in 40% lower values than when actual

operation weighting factors were utilized. It seems that in order to understand methane slip emissions from varying vessel types with different activity profiles, reporting load specific EFs that can then be used together with an activity profile of a certain ship would be most preferable.

Also, it should be noted that the transient conditions such as acceleration and deceleration are not considered in the E3/E2 cycles. In automotive and off-road industry, transient loads and various operating variable conditions are included in the test cycle (e.g. WLTC cycle for EuroVI regulation). Still largely not investigated for marine engines, this topic is expected to gain interest in the future, as transient conditions may significantly degrade engine performance and could produce additional methane-slip.

4.2.2.1 Low load points and mitigation strategies

Increasing methane slip at low engine loads is visible in Figures 6-8 and attention to the phenomenon was also given in [6], [26] where the effect of methane slip at low loads to GWP was discussed and one low-load mitigation strategy investigated. In the study by Sommer et al. [26] emissions were reported for two DF engines of the same engine model, where one of the engines used cylinder deactivation as a strategy to decrease methane slip at low loads. The results from the study are reproduced in Figure 9 where it is shown that cylinder deactivation could markedly reduce the methane slip at low loads. During the study, the engine marked here as LPDF 4-S 2016 (d) used unmodified firing strategy (all cylinders firing at all loads) whereas the engine LPDF 4-S (e) deactivated 3 cylinders at loads below 15%.



Figure 9. Effect of cylinder deactivation on methane slip. Figure reproduced based on [26]

The increased methane slip has been shown to have high impact on the GWP of LNG operated engine at low loads. Peng et al. [6] noted that while methane accounts the greatest fraction of GWP at low loads, its contribution significantly decreases as engine load increases, being largely reduced at load conditions above 75% which underlines the importance of methane slip mitigation at low loads or avoiding operation at low loads entirely. It is understood that often, at low loads, DF engines switch to using liquid fuel as was reported in the study by Anderson et al. [4] at 16% load point. Other mitigation strategies may include avoiding low load operation e.g. by using hybrid propulsion combining engines with batteries or by using shore power at berth as mentioned by Sommer et al. [26].

4.3 Methane slip from engines from different years

Methane emission factors for LPDF 4-S and LPDF 2-S engines as function of engine year are shown in Figure 10 and Figure 11. For LPDF 4-S engines, the reported methane slip EFs for engines from 2016-2019 agree or surpass those from older engines at all engine loads. For the newest engines, the gathered results give twofold trend; the EFs obtained from ship owner data from testbed measurements together with recent on-board study [30] show decreased methane slip compared to older engines whereas results from another recent on-board study [10] show levels exceeding the previous values especially at mid-loads of 25-50%.

For the newest LPDF 4-S engines, ship owner data gives 3.4-4.1 g/kWh at 73-75% load, 2.6-5.9 g/kWh at 40-50% load and 6.6-13.05 g/kWh at 25% load, when the reported on-board values are 3.4-6.3 g/kWh at 75% load, 3.6-16.2 g/kWh at 50% load, and 6.7-70.2 g/kWh at 25% load. Results for engine piloting new combustion concept (marked with year of results published, 2023) show 1.3-1.5 g/kWh at 25-75% loads. Differences in these values affecting the conclusions that can be drawn about engine construction year may originate from operational differences in testbed and on-board as well as different measurement methodology and accuracy as well as comparing engines from different manufacturers which may utilize varving technological solutions.

Considering LPDF 2-S engines (Figure 11), most of the obtained data is from new engines, with one dataset reported for engine from 2019. Again, either ship owner data from on-board measurements or testbed is available, together with the on-board study [10]. Comparing the results to on-board measurement on 2019 engine, the ship owner data again suggests decrease in methane slip with newer engines whereas the results from [10] report increased levels. However, these reported values are in much better agreement with more than 2-fold difference at 23-30% load compared to the case of LPDF 4-S engines where almost 5- to 11-fold difference is observed at low loads.

The increased variability in LPDF 4-S engines compared to LPDF 2-S may partly reflect the higher number of data and different engine manufacturers in case of LPDF 4-S engines. However, the shipowner data for new engines shows that methane slip can be reduced with the newer engine technology and these new data should be considered when the current methane emissions from LNG engines are evaluated.



Figure 10. Methane emission factors as function of engine year for LPDF 4-S engines at 23-20%, 40-50%, and 70-75% loads. Dark tones are used for engines from years 2020-2023, midtones for engines from years 2016-2019 and light tones for engines from years 2015 or older. Measurements conducted in testbed are marked with triangle and labeled with asterisk. If engine year is not available, publication year (indicated by 'publ.')



Figure 11. Methane emission factors as a function of engine year for LPDF 2-S engines at 23-30%, 40-50%, and 70-75% loads. Dark tones are used for engines from years 2020-2022 and midtones for engines from years 2016-2019. Measurements conducted in testbed are marked with triangle and labeled with asterisk.

4.3.1 Comparison of on-board and test-bed measurements

Figure 12 shows the data from Figure 7 grouped by the measurements conducted on-board and in testbed. In their study, Ushakov et al. [20] reported methane emissions from same model LPDF engines in laboratory and at sea and reported a pronounced difference (32%) between on-board measurement compared to manufacturer values. They assumed the difference to be partially explained by differences in engine conditions in laboratory and at sea. Considering all the data for LPDF 4-S engines, reasonable agreement is seen between the values from testbed measurements and those reported from on-board campaigns at 100%, 75%, 50%, and 25% loads. However, no EFs are reported from testbed measurements for loads below 23%. Considering the newest engines, two recent on-board studies show two-fold results, suggesting levels between or below test-bed but also significantly higher values (see Figure 10).



Figure 12. Methane slip emission factor for LPDF 4-S engines measured in testbed (black) or onboard (green). Note logarithmic scale.



Figure 13. Methane slip emission factor for 2-S engines measured in testbed (black) or on-board (purple).

For LPDF 2-S engines (Figure 13), less data is available overall, but it can be noticed that methane slip for <12MW LPDF 2-S engine from 2019 and 11.53 MW LPDF 2-S engine from 2021 [10] show increased methane slip compared to the testbed measurements. However, the on-board results for the LPDF 2-S 2021 of the same size category (>50MW) as the testbed engines agrees well with the testbed results, indicating that in the case of LPDF 2-S data presented here, engine size may have larger impact than the operation environment.

For LBSI engines, testbed and onboard data could be compared only from the study by Ushakov et al. [20] and figure is not reproduced here. Referencing their findings, it was found that testbed and onboard measurement of a same engine model showed rather good agreement at loads above 50% but a discrepancy was observed at lower loads with 400% difference at 25% load. Differences maybe caused due to more challenging engine conditions at sea or comparing engines from different manufacturers. Factors affecting the difference between laboratory and on-board measurements may be operating the engine at stable load without fluctuations, using gas with controlled composition within specification, clean injectors, and combustion chamber whereas changing conditions at-sea may include varying engine loads and conditions [20].

4.4 Other emissions from LNG engines

In addition to methane, information of other gaseous and particulate emission factors (EFs) from LNG engines were collected. It must be noted that only publications which also reported methane slip are included, and thus this section is not a full literature review of other EFs from LNG engines.

EFs of nitrogen oxides (NO_x) were found for 12 LPDF 4-S engines and 8 LBSI 4-S engines. The NO_x EFs varied between 0.5-4.3 g/kWh for LPDF 4-S engines and 0.2-0.4 g/kWh for engine piloting new combustion concept. For LBSI 4-S engines, emissions of 0.26-3.3 g/kWh were found at 25-100% loads, but in two cases, clear load dependency was observed, resulting in EFs of 15-27 g/kWh at 10-25% loads. Majority of the reported LPDF engines achieved the Tier III levels [32] and in the on-board measurements of 2012 and 2016 engines, NO_x emissions for all load points between 25-100% were between 0.5-1.1 g/kWh. In the study of Ushakov et al. [20], overtuning of engines for very low NO_x EFs was discussed as one reason for higher methane slip, underlining the importance of balancing between low NO_x and low methane EFs.

Carbon monoxide (CO)and unburned hydrocarbons are products of incomplete combustion. Peng et al. [6] observed simultaneous increase in CO and hydrocarbons together with methane in conditions where NO_x decreased. In the included literature, CO and hydrocarbon EFs were reported for few LPDF 4-S engines. The CO EFs varied between 1-7 g/kWh at 25-100% load with increased values at lower loads and up to 36.3

g/kWh at idle (0.7-1.8 g/kWh and 12.9 g/kWh at 10% for engine piloting new combustion concept). The emissions for non-methane hydrocarbons (NMHC) were 0.57-2.9 g/kWh, also increasing towards decreasing loads between 100% and 25%, and reaching 20.6 g/kWh at 6% load.

Formaldehyde (HCHO) is a carcinogenic, early interstage product of methane oxidation whose amount typically increases with unburned methane. (CIMAC WG 17, 2014). Peng et al. [6] observed HCHO EFs to increase from 124 to 466 mg/kWh when engine load decreased from 100% to 25% and even further to 2520 mg/kWh at idle. Lehtoranta et al. [31] gave formaldehyde in ppm, but corresponding EFs showed higher values of 210-700 mg/kWh at loads from 85% to 30%. Peng et al. [6], showed that when the same engine ran on diesel, HCHO EFs were 16-32 mg/kWh (loads 100% to 25%) and 337 mg/kWh (idle), pinpointing the importance of considering formaldehyde in addition to methane when developing the LNG engines. Recently, Lehtoranta et al. [30] reported 91-200 mg/kWh at 100 to 25% loads and 545 mg/kWh at 10% load. With engine piloting new combustion concept, 38-54 mg/kWh at 100 to 25% loads and 203 mg/kWh at 10% load were reached.

LNG typically contains very low amounts of sulfur, but in DF engines sulfur dioxide (SO₂) can result from the pilot fuel. No SO₂ EFs for LNG engines were found. Lehtoranta et al. [5] observed <2 ppm concentration for full marine gas oil with reduced sulfur content (<0.001%S) operation and indicated even lower level for DF operation. While SO₂ from LNG engines is low, making them suitable for operation in emission control areas, even low concentrations of sulfur in the exhaust may pose a challenge for the use of methane oxidation catalysts. The use of ultra-low sulfur diesel (ULSD) as pilot fuel is one mitigation option.

Particle number and mass (PN, PM) EFs from LNG engines were included in part of the studies [4]–[6], [29], [30], [33]. Total PM EFs were measured by conversion from particle number size distribution whereas non-volatile PM was measured either by the conversion method or collecting particles on filter. Total PM emissions for the three engines ranged between 0.15-2.6 mg/kWh (10-173 mg/kWh at 6% load), 0.16-0.41 mg/kWh, and 7-11 mg/kWh. Non-volatile PM emissions were 0.13-0.22 mg/kWh, 10 mg/kWh and 20-32 mg/kWh.

Non-volatile particle number emissions were reported for six LPDF 4-S engines. The reported non-volatile PN emissions from different studies were 1.3×10^{11} 1/kWh (particle size cut-off >23nm), $1-1.1 \times 10^{12}$ 1/kWh (>23nm), $1.0-3.0 \times 10^{12}$ 1/kWh (>5.6nm) without notable load dependency in the case of three of the engines. In one case, significant load dependency was observed with non-volatile PN emissions of 1.7-3.0×1012 1/kWh (>6nm) at 50-90% loads being increased by two orders of magnitude to 1.0-3.0×10¹⁵ 1/kWh (>6nm) at low loads of 6-31%. For the remaining engines, moderate load dependency together with increased PN as function of lower cut-point was reported with 0.1-0.4×1012 1/kWh (>23nm) and 0.4-2×1012 1/kWh (>10nm) at 100 to 25% loads and 4×1012 1/kWh (>23nm) and 26×1012 1/kWh (>10nm) at 10% load. For the engine piloting new combustion concept values of 0.7-23×1012 1/kWh (>23nm) and 3-43 ×10¹² 1/kWh (>10nm) were reported. Alanen et al. [34] observed that majority of non-volatile PN from LNG engine may reside well below 23 nm cut-off. In EURO 7 emission regulation considering onroad vehicles, the cut-off point in PN measurement is brought down to 10 nm. For total PN, results for three engines varied with PN EFs in the ranges of 2.3-6.9×10¹² 1/kWh (>5.6nm), 0.18-2.7×10¹⁵ 1/kWh (>6nm), and 3.0-4.0×10¹⁵ 1/kWh (>1nm).

Black carbon (BC) and elemental carbon (EC, analyzed together with organic carbon, OC) EFs were found for LPDF 4-S engines from two studies where they varied between 0.5-1.7 mg/kWh at loads between 25-100% but reached higher values of 5.7-5.9 mg/kWh at 6% load and 6 mg/kWh at idle. Reported together with EC, OC EFs were 2.25-25.3 mg/kWh at loads between 25-100%. However, significant variability with values from 9 to up to 1450 mg/kWh was reported at 6% load and a value of 110 mg/kWh was reported at idle. The OC/EC ratio varied between 1.8-18.3. The BC, EC, and OC emissions from the DF natural gas combustion were generally on a low level as for example Peng et al. [6] observed combustion of ULSD diesel to result in 14.9-38 mg/kWh of EC and 85-151 mg/kWh of OC at 25-100% loads.

Peng et al. [6] further compared the effects of switching a DF marine vessel from diesel to natural gas operation and found that the use of natural gas reduced emissions of NO_x, PM_{2.5}, CO₂, and BC by 92%, 93%, 18% and 97%, respectively. Similarly, Lehtoranta et al. [5] found PM to decrease 72-75% and PNnv by 98-99% when comparing LNG use to marine diesel oil. In a recent review by Aakko-Saksa et al. [3], LNG combustion in DF engines showed significant reduction for PM, but also PN and BC emissions compared to liquid fuels. The reductions on criteria pollutants have been noted as significant in terms of improving air quality in coastal areas. However, in the study of Peng et al. [6], parallel to higher methane emissions, caused by fuel switching, the EFs of CO and formaldehyde increased more than fivefold and sevenfold, respectively. The reduction of PM from natural gas combustion compared to diesel during idling at port was associated with 92% lower cancer risk on the long term, whereas higher formaldehyde emissions from natural gas could remain a concern and possibly require mitigation by catalyst [6].

5 CONCLUSIONS

LNG use as maritime fuel has increased but is still marginal: less than 3% of ships that reported to the EU MRV in 2021 had a carbon factor less than 3.0, which is an indication of alternative fuel use and most likely LNG. The share of LNG of total annual fuel combusted on board ships was 5.9% in 2021 according to the IMO DCS. Methane will be part of future EU mechanisms to cut GHG emissions and mitigation of methane slip is needed to sustain the interest towards the use of LNG as marine fuel.

The amount of available methane slip data is limited with a handful of scientific studies reporting emission factors measured during on-board experiments. LPDF 4-S engines are best represented in the literature, followed by LBSI and LPDF 2-S engines. For HPDF engines, only values originating from manufacturers could be found but methane slip from these engines is considered low.

The emission factors reported for LBSI engines from 2010-2015 showed methane slip of 2.1-25.5 g/kWh at engine loads of 25-100%, indicating higher methane slip towards low loads.

For LPDF 4-S engines, the methane slip from newest engines from 2020-2023 measured in testbed varies between 2.6-4.1 g/kWh at 75-100% load but increased slip of 6.6-13.05 g/kWh is load. From observed for 25% on-board measurements, higher variation of 1.9-6.4 g/kWh at 75-100% loads and even 70.2 g/kWh at 25% have been reported. Differences in these values may originate from operational differences in testbed and on-board as well as different measurement methodology and accuracy as well as comparing engines from different manufacturers which may utilize varying technological solutions. Also, in the case of older engines, increased methane slip at low loads was observed and it remains a concern to be mitigated by the development of engine technology for new engines or by removal via exhaust aftertreatment. It was also shown that cylinder deactivation or new combustion concept can be used to reduce methane slip at low loads.

For LPDF 2-S engines, data could be included for new engines from 2019-2022 and values of 1.9-7.2 g/kWh were found for engine loads between 25% and 100%. Reasonable agreement was found between testbed and on-board studies.

Because load dependency for methane slip is observed, reporting emission factors as function of

engine load instead of weighted emissions over the E3/E2 cycle would be beneficial to understand the methane emissions of ships with varying activity profiles of their engines, as well as for studying the influence of transient load conditions. In their study, Balcombe et al. [10] noted that several continuous emission monitoring systems for methane are commercially available and would enable ships to self-monitor methane, helping further to understand and reduce the emissions. While this paper focuses on Tank-to-Wake emissions, the comparison of different fuel and engine options should consider also Well-to-Wake emissions (e.g. [2], [10], [11]).

6 ACKNOWLEDGEMENTS

The EU HORIZON project GREEN RAY (Grant number: 101056642) is acknowledged.

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