RDE-based assessment of a factory bi-fuel CNG/gasoline light-duty vehicle

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Abstract

On-road exhaust emissions of a Euro 5 factory bi-fuel CNG/gasoline light-duty vehicle equipped with the TWC were assessed considering the Real Driving Emissions (RDE) guidelines. The vehicle was equipped with a Portable Emission Measurement System (PEMS) that enabled the measurement of THC, CO, NO_x, CO₂, and CH₄. With respect to the characteristics of the vehicle, the appropriate Worldwide Harmonized Light-Duty Vehicle Test Cycles (WLTC) were selected and based on the requirements of the RDE legislation a suitable route was conceived. In addition to the moderate RDE-based route, an extended RDE-based route was also determined. The vehicle was driven along each defined route twice, once with each individual fuel option and with a fully warm vehicle.

RDE routes feature a multitude of new driving patterns that are significantly different to those encountered in the NEDC. However, as these driving patterns can greatly influence the cumulative emissions an insight in to local time trace phenomena is crucial to understand, reason and to possibly reduce the cumulative emissions. Original contributions of this paper comprise analyses of the RDE-LDV local time resolved driving emissions phenomena of a CNG-powered vehicle that are benchmarked against the ones measured under the use of gasoline in the same vehicle and under similar operating conditions to reason emission trends through driving patterns and powertrain parameters and exposing the strong cold-start independent interference of CO and N₂O infrared absorption bands in the non-dispersive infrared (NDIR) analyzer. The paper provides experimental evidence on this interference, which significantly influences on the readings of CO emissions. The paper further provides hypotheses why CO and N₂O interference is more pronounced when using CNG in LDVs and supports these hypotheses by PEMS tests.

The study reveals that the vehicle's NO_x real-world emission values of both conceived RDE-based routes when using both fuels are within Euro 5 and type-approval limits. Additionally, the THC and the NMHC emissions of both RDE-based routes using both fuels are within the Euro 5 limits indicating reasonable CH₄ emissions. Notable increases above the type-approval and Euro 5 limits appeared in the CO emissions profile when using gasoline, while the CO₂ emissions profile expectedly also exceeded the type-approval specifications.

Highlights

- **A factory bi-fuel CNG/gasoline LDV was assessed considering the proposed RDE guidelines**
- **CNG RDE-LDV results are benchmarked against the ones of the gasoline fuel**

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- **In addition to THC, CO, NO_x and CO₂, CH₄ was also measured during the RDE test**
- **Strong interference of CO and N₂O, which affects the CO readings of the NDIR is exposed**
- **Hypotheses on CO and N₂O interference for CNG are proposed and supported by PEMS results**

Key words: RDE, PEMS, light-duty vehicle, CNG, gasoline

Abbreviations

CNG – compressed natural gas
RDE – Real Driving Emissions
WLTC – Worldwide Harmonized Light-Duty Vehicle Test Cycles
WLTP – Worldwide Harmonized Light Vehicle Test Procedure
PEMS – Portable Emission Measurement System
LDV – light-duty vehicle
DOC – diesel oxidation catalyst
DPF – diesel particulate filter
EGR – exhaust gas recirculating
SCR – selective catalytic reduction
NEDC – New European Driving Cycle
NTE – not-to-exceed
NDIR – non-dispersive infrared
CF – conformity factor
CADC – Common Artemis Driving Cycle
THC – total hydrocarbons
HC – hydrocarbons
CO – carbon monoxide
NO_x – mono-nitrogen oxides
CH₄ – methane
CO₂ – carbon dioxide
NO – nitric oxide
NO₂ – nitrogen dioxide
N₂O – nitrous oxide
MAW – moving averaging windows
IR – infrared
ppm – parts per million

1. Introduction

Environmental protection is one of the key challenges of modern society and reduction of exhaust emissions from the transport section is one of the crucial pillars to achieve this goal. This aim is clearly reflected in the reduction of emission limits in emission legislations around the globe. However, during the last decades, it also became evident that real-world exhaust emission of the vehicle fleet did not follow the reductions imposed by the emission legislation [11, 13, 14, 18 and 19]. Similar can also be concluded for the fuel consumption [11, 13, 14].

Future emission regulation in the field of light-duty vehicles (LDV) aims to address these challenges relying on the newly established Real Driving Emissions (RDE) legislation [8, 27].

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Additionally, within the framework of the Worldwide Harmonized Light Vehicles Test Procedure (WLTP), a Worldwide Harmonized Light-duty driving Test Cycle (WLTC) is planned to be introduced in 2017 [12, 16, 20, 21]. In EU, it is planned to replace the existing New European Driving Cycle (NEDC) that is in general characterized by lower average and peak engine power requirements and is currently still used for assessing compliance with Euro emissions standards. While the RDE-LDV emission legislation is thoroughly elaborated in the EU [8, 27] and is the main focus of this paper, it must be mentioned that the US will be adopting a new standard named US-EPA Tier 3, which will tighten the emission limits on the established cycles FTP-75, HWFET and the supplemental cycles US06 and SC03. To comply with the new US emission standards, manufacturers will thus have to follow test procedures specified in 40 Code of Federal Regulations (CFR) part 1066 [23, 24].

The introduction of on-road measurements has already proven to be successful for heavy-duty vehicles [29, 30, 31, 32]. The invoking of the Euro VI emission standard for heavy-duty vehicles resulted in severe NO_x and PM reductions during real-world driving [29, 30, 31, 32]. This is primarily due to the application of diesel oxidation catalysts (DOC), diesel particulate filters (DPF), exhaust gas recirculation (EGR), selective catalytic reduction (SCR) and ammonia-selective catalytic reduction (NH₃-SCR) of the exhaust gasses [25, 30, 31, 32].

Although the RDE-LDV test procedure [8, 27] is yet not adopted by the EU Parliament, the documentation is elaborated in detail, and thus allows execution and validation of the tests. The documentation anticipates that, in addition to new vehicles, the test procedure will be conducted on vehicles with mileage up to 100.000 km [9]. For the purpose of testing, the use of portable emission measurement systems (PEMS) and not-to-exceed (NTE) regulatory concepts have been suggested [8, 27]. RDE tests should reproduce a representative range of driving patterns and styles to ensure that the generated emissions correspond to those typically emitted during real driving. The NTE emission limits represent the legal emission limits plus a margin called the conformity factor (CF) [8, 27].

Only a limited amount of studies have addressed the relatively contemporary subject of measuring real-world driving emissions. In [1], two Euro 6 diesel vehicles were evaluated both on-road and over various dyno test cycles. It was stated that real-world driving routes influenced the pollutant emissions [1]. It was further reported that the CO and HC emissions were well within Euro 6 limits for all analyzed routes but NO_x emissions were substantially above the NTE, considering the CF of 1.5 [1]. In [2], the concern of exceedingly high real-world driving emissions was addressed by analyzing gaseous emissions of one Euro 6 and six Euro 4-5 diesel vehicles with PEMS. Although considerably lower NO_x emissions of the Euro 6 vehicle compared to the Euro 4-5 vehicles were observed, it was still discovered that all tested vehicles substantially exceeded their respective NO_x emission limits [2]. In [3], the Association for Emissions Control by Catalyst (AECC) published their findings on a series of conducted tests on a Euro 5 gasoline vehicle and on a Euro 6 diesel vehicle. Substantial differences for some pollutants between RDE emissions measured with PEMS equipment compared to the dynamometer tests using NEDC and CADC cycles were reported in this analysis. The study revealed that both vehicles emitted significantly higher values of NO_x on the RDE route than on the dynamometer cycles, which also exceed their respective Euro limit. Moreover, it was claimed that the high NO_x emissions particularly occur under high velocities and loads [3], which is expected. Nevertheless, RDE emissions of CO and HC were below the respective Euro limit values for both vehicles. The International Council of Clean Transportation (ICCT) published an analysis of on-road emissions data from several Euro 6 passenger vehicles in [4]. It was reported that the CO and THC emissions were well below the Euro 6 emission limits, while the NO_x real-world emissions exceeded the regulatory emission limit by more than six times. A similar conclusion was made in [54], where the German Ministry of Transport

commissioned a series of laboratory and on-road vehicle tests of 53 different vehicle models in order to determine the use of illegal defeat devices by major vehicle manufacturers. In [5], the Joint Research Centre analyzed on-road emissions of 12 light-duty diesel and gasoline vehicles that comply with Euro 3-5 emission limits. It was disclosed that the NO_x emissions of diesel vehicles, including Euro 5 vehicles, notably exceeded the respective Euro 3-5 emission limits [5]. On the other hand, NO_x emissions of gasoline vehicles as well as CO and THC emissions of both diesel and gasoline vehicles generally remain within Euro 3-5 emission limits.

In [15], a bi-fuel Euro 5 (CNG/gasoline) vehicle was tested over the NEDC on a chassis dynamometer. It was noted that the CO₂ emissions were 24-25% lower when running on CNG compared to the gasoline use [15], which is in line with H/C ratio of both fuels based on the energy content. In [15], it was also stated that emission testing of CNG under real-world conditions, which is not presented in [15], is necessary in order to have a complete picture of the environmental performance of CNG usage in Euro 5/6 vehicles. In [22], steady-state tests on a chassis dynamometer were performed with a passenger car powered by a retrofitted spark ignition engine using gasoline and CNG. The paper reports that the CO emissions when using CNG were significantly lower compared to the ones when using gasoline [22]. In addition, it was reported that the vehicle emitted more NO_x when using CNG than when using gasoline. It was concluded that higher NO_x emissions are the main concern when using CNG [22]. Based on the literature review it can thus be concluded that there is a need for analyses of the RDE-LDV emissions of a CNG-powered vehicle, which is the subject of the present paper, as such type of analyses were, to the best of author's knowledge, not yet published. This paper presents analyses of the time resolved on-road real driving emissions phenomena of a CNG-powered vehicle, benchmarking them with those measured for gasoline in the same vehicle under similar operating conditions and reasoning of the emission trends through driving conditions and powertrain parameters. The latter is of particular importance as this topic is not deeply explored for RDE-based cycles and thus interrelation of driving conditions and powertrain parameters to the time resolved emission traces provide insightful information that can contribute to reducing cumulative emissions. To additionally strengthen the paper, an indicative comparison and assessment of real-world on-road emissions of a Euro 5 bi-fuel CNG/gasoline light commercial vehicle equipped with the TWC to the Euro 5 regulatory limit being the type-approval limit for the tested vehicle is presented. The deviation of realistic on-road emission footprint along RDE-based routes from the type-approval limit serves an important information of the vehicle's performance over the plausible vehicle operation range. This was recently mostly done for diesel and gasoline vehicles but rarely for CNG vehicles.

It is also important to note that the Euro 5 introduced stricter methane limits for positive ignition vehicles, specifically with enforcing an NMHC (non-methane hydrocarbons) limit which complements the THC (total hydrocarbons) limit. To address this concern, this paper reports CH₄ emissions measured in real-world driving. This was possible with the application of a new generation PEMS that is, besides measuring CO, CO₂, NO, NO₂ and THC emissions, also capable of measuring CH₄ emissions. This further strengthens this study, as, to the best of our knowledge, CH₄ emissions were not yet measured during the real-world driving.

Moreover, the paper additionally exposes the strong interference of CO and N₂O infrared absorption bands, which can significantly affect the CO readings from the non-dispersive infrared (NDIR) sensor being the most widespread sensor in a PEMS. This hypothesis was made by analyses of time traces of the CNG fuel vehicle and it was proven by an independent experiment with a similar commercially available analyzer of another manufacturer that clearly exposed correlation between N₂O presence in the gas fed to the NDIR analyzer and the CO readings. Since the CO measurement error appears only when the vehicle was using CNG, the paper also suggests a plausible route of N₂O formation considering engine-out emission and

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chemical kinetics in the TWC specific to the use of CNG. Consequently, the presence of N₂O can significantly influence the test cycle cumulative CO emission readings. Due to the clear physical background of these phenomena, it can be considered that this is a general challenge rather than an equipment specific effect.

The RDE regulation opens a huge variation space in terms of driving patterns that are significantly different to the ones encountered in the NEDC. As one of the main aims of the paper is focused on analyzing these, previously unexplored, driving patterns and to provide a cause and effect explanation between driving patterns, powertrain parameters and tailpipe emissions, we purposely kept the number of additional variations to a minimum. Therefore, considering complex TWC related phenomena during the catalyst light-off phase, cold starts were intentionally omitted to ensure clarity of analyses, whereas other RDE trip boundary conditions were respected.

To the best of the author's knowledge, this paper comprises the following original contributions: a) analyses of the RDE-LDV emissions of a CNG-powered vehicle; b) benchmarking of the CNG RDE-LDV results with the ones measured for gasoline in the same vehicle and under similar operating conditions; c) measurements of on-road RDE-based route CH₄ emissions using a PEMS; d) exposing the strong interference of CO and N₂O infrared absorption bands, which can significantly influence the CO readings from the non-dispersive infrared sensor (NDIR); and e) revealing the hypotheses why CO and N₂O interference is more pronounced when using CNG in LDVs.

2. Experiment

2.1. Test vehicle and PEMS instrumentation

Table 1 summarizes key characteristics of the vehicle. Test vehicles' payload comprised of the driver and a PEMS operator who also served as a witness to the test and the test equipment including the power supply devices, summing up to 1996 kg.

Table 1. Test vehicle's characteristics

Engine capacity [cm ³]	1984
Power output	80 kW@5400 RPM
Compression ratio	13.5: 1
Emission standard	Euro 5
Unladen weight [kg]	1580
Transmission	Manual
Certification CO emissions using CNG[g/km]	0.3768
Certification HC emissions using CNG [g/km]	0.0294
Certification NO _x emissions using CNG [g/km]	0.0168
Certification CO ₂ emissions using CNG [g/km]	156
Odometer at start of tests [km]	11202
CNG fuel capacity [kg]	26
Gasoline fuel capacity [liters]	13

A Portable Emissions Measurement System (PEMS) was used to measure emissions during on-road driving over pre-selected routes. The system comprised of an AVL M.O.V.E. PEMS equipment coupled with an exhaust flow meter (EFM). The PEMS enabled the measurement of THC, CO, NO_x, CO₂, and CH₄. An additional feature of the used PEMS is that analyzers of all the aforementioned emissions are combined within one device.

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The PEMS analyzer and other main components were installed in the cabin of the vehicle, while the EFM, GPS antenna, and the weather station were installed outside of the vehicle. The EFM was attached to the vehicle's tailpipe and its sensors placed according to the test procedure's specifications. The exhaust mass flow was determined based on a precisely measured differential and absolute pressure, on the exhaust temperature measured with a K-type thermocouple and with a calibration based equation. Along with the aforementioned parameters, the OBD information was also logged. All gaseous emissions and vehicle performance parameters were sampled at a frequency of 1 Hz.

The gasoline used during the measuring campaign was an EN228 standard 95-octane gasoline fuel. The composition of CNG used during the measuring campaign is given in Table 2. The CNG composition presented in Table 2 indicates that the used CNG consisted mostly of methane (CH₄). The test sequence of the measuring campaign is presented in Table 3.

Table 2. CNG composition during the measuring campaign

Component	Average value [mol %]
Methane	96.170
Butane	2.307
Propane	0.519
i-Butane	0.088
n-Butane	0.084
neo-Pentane	0.000
i-Pentane	0.018
n-Pentane	0.011
Hexane+	0.015
Nitrogen	0.583
Carbon dioxide	0.206

Table 3. Measuring campaign test sequence

Day [/]	Driven route, fuel [/]
Day 1	WLTP, CNG
Day 2	RDE Moderate route, gasoline
Day 2	RDE Moderate route, CNG
Day 3	RDE Extended route, gasoline
Day 3	RDE Extended route, CNG
Day 4	Other routes, CNG

2.2. WLTP

According to the Annexes laid down in the Global Technical Regulation no. 15 (GTR.15) [7], several conditions of executing the WLTP must be met. The first step in the WLTP is to execute the WLTC considering the category of the testing vehicle. As indicated in the Introduction, the cold start phase was conscientiously neglected. According to the test procedure of Annex 1 [7], the ratio of the unladen mass of the vehicle to its power and vehicle's maximum velocity suggests that the vehicle resides in the WLTP Class 3 category. The maximum velocity of the test vehicle exceeds 120 km/h, therefore, the vehicle settles in WLTP Class 3b. A complete WLTC cycle for a Class 3b vehicle consists of a low phase (Low₃),

medium phase (Medium₃₋₂), a high phase (High₃₋₂) and an extra high phase (Extra High₃) velocity as presented in Fig. 1.

Fig. 1. The WLTC velocity trace for a Class 3b vehicle [7]

In addition to the RDE-based routes, WLTC cycles were also driven on-road in the presented analysis. The WLTC was driven on-road with CNG fuel only. The measuring campaign was performed at night on a straight road with the altitude difference between the start and the end point being roughly 3m. For the purpose of this campaign, the road was closed, therefore, no traffic was present. The average mean squared error between the legislative vehicle velocity and the on-road driven vehicle velocity was 2.8%. The authors of this study find this error to be low enough to produce a plausible basis for comparative analyses, as the main goal of this study is to present and compare fuel and driving condition specific phenomena. Considering the results published in [17] where the vehicle with an almost identical engine was driven over the NEDC with CNG, gasoline, and LPG fuels a fair estimate of gasoline WLTC CO₂ emissions can be made from the CNG WLTC CO₂ emission profile given in section 3.1 by considering H/C ratio of both fuels based on the energy content.

2.3. RDE

The RDE test program was designed considering the 1st package of the RDE-LDV legislation published in March 2016 [8] and the 2nd package of the RDE-LDV legislation published in April 2016 [27]. A general overview of the RDE and WLTP background is given in Appendix A. The RDE performance should reflect the vehicle emissions on the road, operated over its normal driving patterns, conditions, and payloads [8]. In order to evaluate the RDE emissions over the entire plausible vehicle operation range, two RDE-based test routes were developed; one considering the RDE moderate conditions (hereinafter referred to as RDE Moderate route) and the other considering the RDE extended conditions (hereinafter referred to as RDE Extended route). As indicated in the Introduction, given that the cold start condition

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was not considered when driving the designed routes, the terminology “RDE-based” is used for the analyzed cycles, as the rest of the RDE trip boundary conditions were respected. Therefore, a significant effort was put in maintaining a warm TWC in order to rule out any effect of cold start, which is known to significantly influence the TWC conversion efficiency. Fig. 2 presents the on-road visualization of both driven routes.

The main objective of the RDE Moderate route is to represent a conventional RDE test that is planned to be performed by the appropriate technical services [29] with the invoking of the Euro 6/d standard while the main objective of the RDE Extended route is to test the vehicle's emission footprint on a more power-demanding route. The conditions become extended when at least one of the ambient (temperature or altitude) boundary condition is extended [8]. The legislation states that the maximum altitude above sea level for a moderate route is 700m while the maximum altitude above sea level for an extended route is 1300m [8]. RDE Extended route was developed in a manner that the maximum altitude above the sea level condition is extended, therefore it must reside between 700m and 1300m [8], which can be seen from the data in Table 4 presenting the specifications of RDE Moderate and RDE Extended route using both fuel options compared to the RDE trip boundary conditions. The data comparison in Table 4 also reveals that all of the main RDE legislation specific trip boundary conditions were respected throughout the measuring campaign. The test vehicle was driven twice over both developed routes, once with gasoline fuel and once with CNG.

The verification procedure was done considering the EMROAD (also known as the Moving Averaging Windows) method laid down in Appendix 5 of the 1st package of the RDE legislation [8].

Fig. 2. RDE-based routes: red line - RDE Moderate route, green line - RDE Extended route

Table 4. Specifications of RDE Moderate and RDE Extended route using both fuel options compared to the main RDE legislation trip boundary conditions

	RDE Moderate route		RDE Extended route		Legislation boundary conditions
	Gasoline	CNG	Gasoline	CNG	
Trip duration [min]	107	108	102	95	90 - 120
Trip distance [km]	98.51	98.62	90.33	90.26	> 48
Urban route share distance [km]	42.9	41.04	45.38	43.6	> 16
Rural route share distance [km]	32.92	32.81	20.82	22.38	> 16
Highway route share distance [km]	22.69	24.77	24.13	24.28	> 16
Urban route share trip average speed [km/h]	39	37	37	40	15 - 40
Rural route share trip average speed [km/h]	71	71	72	72	60 – 90
Highway route share average speed [km/h]	109	111	120	122	100 - 145
Cumulative positive altitude gain [m]	400	400	915	915	< 1200
Maximum trip altitude [m]	404	404	860	860	Moderate < 700 Extended < 1300
Altitude difference [m]	8	8	82	82	< 100

3. Results

Emission results analysis of the WLTC test, the RDE Moderate route, and the RDE Extended route is presented in this section.

First, cumulative emissions of the complete data set for each route using both fuels are presented. This data is then compared to the corresponding Euro 5 emission limits and type-approval limits in an attempt to initially estimate the vehicles' emission footprint. This indicative comparison against the regulatory limit serves as a good indicator to assess the vehicle's on-road emission footprint along an RDE-based route, which is much more power-demanding than the NEDC and symbolizes a plausible real-life driving route.

Secondly, as this paper is focused on the EMROAD method of RDE verification, which is based on moving averaging windows (MAW), an analysis that covers the results of windowed mass emissions of CO, THC, NO_x and CO₂ is presented. Such an analysis is helpful in terms of a comprehensive analysis of the gathered emissions data. Cumulative emissions of THC, CO, and NO_x over each window are compared to the corresponding Euro 5 emission limits while cumulative CO₂ emissions are compared to the type-approval CO₂ emission values over NEDC.

As a window analysis alone cannot explain individual phenomena in the emission traces there is a need for a more detailed analysis. Hence the third analysis in this section covers the instantaneous emission traces. This analysis is performed with a high temporal resolution, which allows a more accurate insight into time-resolved phenomena of the emissions trace. Instantaneous emission results of CH₄, THC, CO, and NO_x are presented in conjunction with the corresponding exhaust gas temperature and mass flow measurements, which is beneficial as it provides an insight into the thermal history of the engine and the aftertreatment system. Additionally, the aforementioned results are amended by traces of velocity, altitude and the air-

to-fuel ratio (λ). Instantaneous velocity provides a useful characterization of the driving pattern, whereas the air-to-fuel ratio provides an insight into the composition of the in-cylinder charge and thus of exhaust emissions. The altitude reflects the road gradient, which in combination with the velocity and the accelerometer reading provides an insight into the power demand of the vehicle. For analysis purposes, the instantaneous emission results, except CO emissions, of each route using both fuel options will be presented on the same scale and with the same accuracy level.

3.1. WLTC test results

In order to minimize the influence of the potential road gradient and wind, the WLTC cycles were driven twice on the same flat road, once in every direction. Emissions of both test runs were averaged. Measurement results of each WLTC section are presented in Table 5. The most interesting data for the RDE evaluation is the cumulative WLTC CO₂ mass value, which in the analyzed case sums up to 5072 g. According to the EMROAD procedure, half of this value is used as CO₂ MAW reference value used for RDE verification of routes driven with CNG.

Table 5. WLTC emissions of individual WLTC sections (presented in Fig. 1) and cumulative WLTC emissions with CNG

	Low	Medium	High	Extra High	Cumulative
Gas	Mass [g]	Mass [g]	Mass [g]	Mass [g]	Mass [g]
CH ₄	$1.9 \cdot 10^{-3}$	$5.8 \cdot 10^{-3}$	$10.2 \cdot 10^{-3}$	$46.7 \cdot 10^{-3}$	$64.6 \cdot 10^{-3}$
CO	1.447	1.926	1.880	2.136	7.389
CO ₂	740.4	1044	1405	1882	5072
NMHC	$2.8 \cdot 10^{-3}$	$2.1 \cdot 10^{-3}$	$2.4 \cdot 10^{-3}$	$0.6 \cdot 10^{-3}$	$7.9 \cdot 10^{-3}$
NO	0.046	0.047	0.102	0.302	0.497
NO ₂	$10.2 \cdot 10^{-3}$	$5.3 \cdot 10^{-3}$	$14.4 \cdot 10^{-3}$	$10.8 \cdot 10^{-3}$	$40.7 \cdot 10^{-3}$
NO _x	0.056	0.052	0.116	0.313	0.537
O ₂	101.1	82.82	124.4	121.3	429.6
THC	$4.7 \cdot 10^{-3}$	$7.9 \cdot 10^{-3}$	$12.6 \cdot 10^{-3}$	$47.3 \cdot 10^{-3}$	$72.5 \cdot 10^{-3}$

In [17], a vehicle with an almost identical SI engine and comparable mass was tested and it was reported that the engine emitted 27.88% more CO₂ when using gasoline than when using CNG. Therefore, it can be estimated that cumulative WLTC CO₂ emissions of the vehicle analyzed in this paper while using gasoline equal approximately 127.88 % of WLTC CO₂ emissions while using CNG thus yielding a value of 6486 g.

3.2. Emissions of the RDE Moderate route

Table 6. Real-world driving emissions using CNG and gasoline compared to type-approval and Euro 5 emission limits for the RDE Moderate route

	CNG	Gasoline	Type-approval	Euro 5 limit
M_{CO} [mg/km]	164.37	3474.2	376.8	1810
M_{THC} [mg/km]	4.16	12.6	29.4	130
M_{NMHC} [mg/km]	0.38	4.07	N.A.	68
M_{NO_x} [mg/km]	11.73	5.88	16.8	75
M_{CO_2} [g/km]	166.91	260.28	156	unregulated
M_{CH_4} [mg/km]	3.78	8.58	N.A.	unregulated

M_{NO} [mg/km]	9.85	5.12	N.A.	unregulated
M_{NO_2} [mg/km]	1.88	0.75	N.A.	unregulated

Table 6 presents the real-world driving emissions using CNG and gasoline compared to type-approval and Euro 5 emission limits for the RDE Moderate route using CNG and gasoline. Results in Table 6 indicate that the THC and NO_x emissions when using both fuel options are within the type-approval and Euro 5 limits. Additionally, it can be seen that the NMHC emissions when using both fuel options are within the Euro 5 emission limits. The CO emissions when using gasoline are nearly a factor of two higher than the Euro 5 emission limits and a factor of nine higher than the type-approval limits, while the CO emissions when using CNG are within the type-approval and Euro 5 emission limits. The CO_2 emissions when using both fuel options exceed the type-approval limit. Reasoning and insight into these results will be elaborated in the following sections.

3.2.1. Window analysis

Fig. 3. Evaluation of windowed emissions compared to Euro 5 emission limits and type-approval CO_2 values for RDE Moderate route using a.) CNG and b.) gasoline fuel

Fig. 3 presents an evaluation of windowed emissions compared to Euro 5 emission limits and type-approval CO_2 values for the RDE Moderate route. From Fig. 3a, it can be seen that emissions of CO, THC, and NO_x when using CNG of all individual windows are below Euro 5 limit confirming the results in Table 6. Almost 20% of individual windows recorded CO_2 emissions above type-approval value. From Fig. 3b, it can be seen that roughly 30% of individual windows record CO emissions above Euro 5 limit when using gasoline. THC and NO_x emissions of all individual windows when using gasoline are below Euro 5 limit. CO_2 emissions results from Table 6 when using gasoline are explained by the fact that almost 60% of individual windows record CO_2 emissions above type-approval, which is expected as the vehicle was type-approved using CNG.

The instantaneous emission analysis presented in Section 3.2.2 provides more detailed reasoning of the time intervals of the RDE Moderate route at which the individual windows recorded above type-approval/Euro 5 limit values of the measured emissions.

3.2.2. Instantaneous emission analysis

Fig. 4. Instantaneous on-road emissions of RDE Moderate route using CNG: a) velocity and altitude profile, b) exhaust gas mass flow and temperature, c.) CH_4 emissions and λ , d.) THC emissions, e.) CO emissions, f.) NO_x emissions

Fig. 4 summarizes the most relevant time traces of the vehicle, engine and emission parameters that are required to establish interrelation between listed parameters and to reason observed emission trends for the RDE Moderate route using CNG. In Fig. 4c and 4d, it can be observed that the shape of time traces of the CH₄ and THC emission quantitatively coincide, which is expected. Peaks of both emissions until 1500s are mainly a consequence of sudden load decreases and increases combined with corresponding temperature fluctuations around the catalyst temperature being at the edge of the light-off temperature, which is indicated by the time trace of the exhaust gas (EG) temperature in Fig. 4b. The CH₄ and THC emissions feature higher values also in the time interval between 3400s and 4500s, which correlates to the downhill driving (Fig. 4a). In this regime, low engine loads lead to the reduction of the catalyst temperature, hindering a successful conversion in TWC. Further peaks of both emissions can be observed between 5000s and 6000s. These emission peaks primarily correlate with rapid load and consequently EG mass flow increases seen in Fig. 4b. Furthermore, within this time window, the engine power frequently exceeds maximum engine power attained during the NEDC cycle since, as discernible in Fig. 4a. In this time window, the vehicle was operated on the motorway with varying vehicle velocity and road gradient. This indicates a potential CH₄ and THC breakthrough, as engine power and thus velocities in the TWC in these operating regimes exceeded the maximum engine power attained during the NEDC cycle being, in general, a sizing guideline for the TWC. Further effects that contribute to these CH₄ and THC emission trends can be summarized as retarded ignition timing to prevent knock and increased engine speed. Both effects namely contribute to higher engine-out CH₄ and THC emissions.

From the comparison of Fig. 4c and Fig. 4d, it can be observed that CH₄ emissions account for most of the THC emissions, which is in agreement with the results in Table 6. This is also an expected trend considering the fuel composition, whereas this further reasons relatively high TWC temperatures that are required to attain high conversion of the CH₄ [26, 36].

In Fig. 4e two types of CO emission peaks can be observed. The first type of CO peaks is characterized by rapid increases and decreases of the CO emission that correlate to THC peaks and are observed during motorway driving at high engine loads (in the time interval around 5000s). The reasoning of these realistic CO emission peaks is thus similar as of the ones for the THC emission during the motorway driving. The second type of CO emission peaks, which is observed much more often, is associated with the rapid rise following with an exponential fall of the CO emission and can be seen throughout the entire RDE-based route. The unusual shape of these peaks, their very long decay times and missing correlation of their occurrence with the relative air-to-fuel ratio (λ) or temperature and velocity in the TWC, lead to the hypothesis that these peaks are a misconstruct generated by the NDIR analyzer. The reasoning of this falsity is explained with the strong interference of CO and N₂O infrared absorption bands as elaborated in Section 3.4 and Appendix C, whereas, the paper also suggests a plausible route for N₂O formation considering engine-out emissions and chemical kinetics in the TWC specific to the use of CNG being elaborated Section 3.4 and in Appendix B.

When analyzing NO_x emissions, it is worth mentioning that when using CNG the engine does not enrich the combustion mixture to λ under 1, which combined with the earlier ignition causes high cylinder temperature, both favoring NO_x formation. Instantaneous NO_x emissions are presented in Fig. 4f. NO_x emission peaks are mostly a consequence of sudden load decreases and increases. They mostly coincide with lean air-to-fuel ratios (λ), which can be seen throughout the entire time interval of the RDE, since the conversion efficiency of NO_x in a TWC is considerably reduced at lean operating conditions [33]. In addition, NO_x emission peaks until 5000s can also be reasoned with lower TWC temperatures being at the edge of the light-off temperature. Lower TWC temperatures can be argued with the driver being less aggressive and the RDE Moderate route proving to be on the lower bound of the vehicle power

demand in the first 5000s of the RDE Moderate route. Higher NO_x peaks from 5000s to 6000s are associated with higher loads, indicated by high EG mass flows and temperature. Since the EG temperature chart illustrated in Fig. 4b indicates sufficiently high TWC temperatures for successful NO_x conversion, the higher NO_x emissions peaks within this time window suggest a potential NO_x breakthrough and consequently a similar interpretation for higher emissions after TWC as given for the CH_4 and THC emissions within this time frame.

Fig. 5. Instantaneous on-road emissions of RDE Moderate route using gasoline: a) velocity and altitude profile, b) exhaust gas mass flow and temperature, c.) CH₄ emissions and λ , d.) THC emissions, e.) CO emissions, f.) NO_x emissions

Fig. 5 summarizes the most relevant time traces of the vehicle, engine and emission parameters that are required to establish interrelation between listed parameters and to reason observed emission trends for the RDE Moderate route using gasoline. In Fig. 5c and 5d, it can be seen that the shape of time traces of the CH₄ and THC emission coincide, but in contrast to the CNG use where THC emissions consist primarily of CH₄ being mainly unburned fuel, it can be seen that peaks of the THC emissions are significantly higher than those of the CH₄ emissions using gasoline, where CH₄ is mainly formed during the combustion process from higher hydrocarbons. The results in Table 6 further support this claim. When analyzing both traces, it can be observed that the CH₄ and THC emission peak at the start of the route is a consequence of a sudden load increase combined with low conversion rates in the TWC. Notable CH₄ and THC emission peaks do not emerge until the start of the uphill (in the time interval around 2600s) where a rapid increase in vehicle velocity and road grade is detected, denoting high load and rich mixture first yielding more engine-out CH₄ and THC emissions due to a lack of oxygen and further hindering their conversion efficiency in the TWC [33]. In addition, during this high load operating regime using gasoline, spark timing is further retarded compared to the use of CNG due to relatively high compression ratio designed for use of the CNG. It can be seen that higher CH₄ and THC emission peaks re-emerge at 5000s, roughly when the vehicle entered the motorway. Within the time window of motorway operation (between 5000s and 6000s) the engine frequently exceeds maximum engine power attained during the NEDC cycle indicating rich mixture phases combined with retarded spark timing as well as potential CH₄ and THC breakthrough. Apart from the aforementioned sections of the RDE-based route where the vehicle was under high load and operated with below-stoichiometric λ , it can be seen that the emissions of CH₄ and THC during the rest of the RDE-based route were nearly negligible, denoting good performance of the engine and the TWC.

Instantaneous CO emissions are presented in Fig. 5e. It can be seen that the CO emissions are mainly characterized by the trend of λ and thus rich mixture, which is expected as the lack of oxygen and retarded spark timing yield more engine-out CO emissions and their subsequent lower conversion efficiency in a TWC [33]. The CO emission peaks hence reach higher values within the time intervals of higher loads (the ascent between 2500s and 3000s and motorway operation between 5000s and 6000s). It is important to note that, in contrast to CO emissions traces when using CNG as a fuel, the CO emissions traces when using gasoline do not feature the presumably false CO peaks characterized by the rapid rise following with an exponential fall and missing correlation with the relative air-to-fuel ratio (λ) or temperature and velocity in the TWC. Additionally, the CO emission traces were inspected with a roughly 20x magnification to account for higher display scales of CO emission traces when using gasoline compared to when using CNG. It can be reported that no CO measurement errors nor any indication of CO measurements errors being masked by the higher CO scale were observed. Therefore, it can be assumed with high certainty that the measuring error detected when using CNG is not present for the use of gasoline.

Instantaneous NO_x emissions are presented in Fig. 5f. It can be seen that the NO_x emission peaks from 0s to 5000s mostly correlate with lean air-to-fuel ratios (λ) and with low TWC temperatures being at the edge of the light-off temperature. Similar to the NO_x peaks between 5000s and 6000s when using CNG, it can be seen that the NO_x peaks within this time interval are also associated with higher loads when using gasoline, indicating a potential NO_x breakthrough. Furthermore, it is important to note that lower cumulative values of the NO_x

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emissions when using gasoline in general result from lower engine-out NO_x emissions being mainly influenced by rich mixture and retarded spark timing both reducing engine-out NO_x emissions and further by their higher conversion efficiency in TWC due to high CO and low or nearly negligible O_2 concentration promoting NO reactions with CO.

3.3. Emissions of the RDE Extended route

Table 7. RDE Extended route emissions using CNG and gasoline compared to type-approval and Euro 5 emission limits

	CNG	Gasoline	Type-approval	Euro 5 limit
M_{CO} [mg/km]	227.82	4385.6	376.8	1810
M_{THC} [mg/km]	18.20	12.96	29.4	130
M_{NMHC} [mg/km]	3.14	2.38	N.A.	68
M_{NO_x} [mg/km]	15.50	8.89	16.8	75
M_{CO_2} [g/km]	212.52	289.22	156	unregulated
M_{CH_4} [mg/km]	15.06	10.58	N.A.	unregulated
M_{NO} [mg/km]	14.15	5.69	N.A.	unregulated
M_{NO_2} [mg/km]	1.34	3.19	N.A.	unregulated

Table 7 presents the real-world driving emissions when using CNG and gasoline compared to type-approval and Euro 5 emission limits for the RDE Extended route when using CNG and gasoline. Results in Table 7 indicate that the THC and NO_x emissions when using both fuel options are within the type-approval and Euro 5 limits. Additionally, it can be seen that the NMHC emissions when using both fuel options are within the Euro 5 emission limits. The CO emissions when using gasoline are nearly 2.5 times higher than the Euro 5 emission limits and nearly 12 times higher than the type-approval limits, while the CO emissions when using CNG are within the type-approval and Euro 5 limits. The CO_2 emissions when using both fuel options exceed the type-approval limit, which is obvious due to the driving pattern. Reasoning and insight into these results will be elaborated in the following sections.

The comparison of RDE Moderate route emissions given in Table 6 to the RDE Extended route emissions presented in Table 7 reveals that when using CNG values of all of the monitored emissions except NO_2 have notably increased. A similar comparison for the gasoline fuel reveals that THC and NO emissions feature similar values for both routes, NMHC emission feature significant decrease for the RDE Extended route, while all other emissions have notably increased for the RDE Extended route. Compared to the RDE Moderate route when using CNG it can be seen that the CO_2 emissions of the RDE Extended route when using CNG have risen by 27%, whereas the CO_2 emissions when using gasoline have risen by only 11 %. This phenomenon can be explained by the decrease in the difference of the retarded ignition timing between using CNG and gasoline in the RDE Extended route compared to the RDE Moderate route.

3.3.1. Window analysis

Fig. 6. RDE Extended route evaluation of windowed emissions compared to Euro 5 emission limits and type-approval CO₂ values, a.) using CNG, b.) using gasoline

Fig. 6 presents the RDE Extended route evaluation of windowed emissions compared to Euro 5 emission limits and type-approval CO₂ values. From Fig. 6a it can be seen that emissions of CO, THC, and NO_x when using CNG of all individual windows are below Euro 5 limit which confirms the results in Table 7, whereas almost 50% of individual windows recorded CO₂ emissions above type-approval value. From Fig. 6b it can be seen that roughly 30% of individual windows record CO emissions above Euro 5 limit while roughly 1% of individual windows record THC emissions above Euro 5 when using gasoline. NO_x emissions of all individual windows when using gasoline are below Euro 5 limit, while CO₂ emissions results from Table 7 are explained by the fact that almost 50% of individual windows record CO₂ emissions above type-approval.

Analyses of instantaneous emissions are presented in Section 3.3.2 and provide more detailed reasoning of the time intervals of the RDE Extended route at which the individual windows recorded above type-approval/Euro 5 limit values of the measured emissions.

3.3.2. Instantaneous emission analysis

Fig. 7. Instantaneous on-road emissions of RDE Extended route using CNG: a) velocity and altitude profile, b) exhaust gas mass flow and temperature, c.) CH₄ emissions and λ , d.) THC emissions, e.) CO emissions, f.) NO_x emissions

Fig. 7 summarizes the most relevant time traces of the vehicle, engine and emission parameters that are required to establish interrelation between listed parameters and to reason observed emission trends for the RDE Extended route using CNG. The reasoning of the phenomena causing emission peaks of various gaseous emissions is similar to the interpretation laid down in section 3.2.2 (RDE Moderate route when using CNG), therefore, this section will focus on explaining the phenomena, seen only in the RDE Extended route when using CNG.

In contrast to RDE Moderate route, the RDE Extended route features a longer downhill leading to higher CH₄ and THC emissions. Within the time interval between 2600s and 3000s, when the vehicle was descending, the engine operated in general at very low and predominantly with a lean mixture yielding high CH₄ and THC emission. Additionally, Fig. 7b indicates that the EG temperature was remarkably reduced in this section, suggesting low conversion rates in the TWC [26, 36]. Larger spark advance produces higher combustion temperatures and lower exhaust temperatures. The latter combined with a very high methane share in the THC emissions of exhaust gasses and lower efficiency of the THC conversion in the TWC due to its faster degradation and low exhaust gas temperatures results in higher THC emissions.

Low CO emissions within the time interval between 2600s and 3000s where other pollutants peaked are reasoned with high combustion temperatures since more CO is converted to CO₂ at higher temperatures. The CO emissions of a CNG vehicle are typically low since the vehicle operates with stoichiometric ratio and typically does not enrich the combustion mixture below $\lambda=1$. In relation to the rest of the CO emission traces, it is important to note that two types of CO emission peaks are observed also for the RDE Extended route (Fig. 7e). Hence, analysis of CO emissions of both RDE-based routes suggests a similar issue with CO emission measurement readings when using CNG.

In Fig. 7f it can be seen that the NO_x emission peaks, in general, reach similar ppm values compared to the NO_x emission peaks of the RDE Moderate route when using CNG. Exceptions, seen as higher NO_x emission peaks occurring at 650s, 1550s and 4700s are a consequence of a rapid load increase yielding power outputs of the engine well above the NEDC range leading to the previously explained chain of events. The NO_x emissions peak at 2800 s when the vehicle was descending is a consequence of a load increase at lean operating conditions leading to NO_x breakthrough due to low EG temperatures and excess oxygen.

Fig. 8. Instantaneous on-road emissions of RDE Extended route using gasoline: a) velocity and altitude profile, b) exhaust gas mass flow and temperature, c.) CH₄ emissions and λ , d.) THC emissions, e.) CO emissions, f.) NO_x emissions

Fig. 8 summarizes the most relevant time traces of the vehicle, engine and emission parameters for analyses of time-resolved traces when using gasoline. Again, this section will focus on explaining the phenomena, seen only in the RDE Extended route when using gasoline.

Instantaneous THC emission can be observed in Fig. 8d. The RDE Extended route features a steeper and more power-demanding uphill, which explains higher THC emissions compared to the THC emissions of the RDE Moderate route in the uphill section. Apart from the ascent (between 1700s and 2300s) and the motorway section (between 3800s and 4700s), the results indicate negligible THC emissions on the rest of the RDE Extended route. The main cause of high THC emissions is again rich mixture and retarded spark timing as well as all resulting phenomena already explained previously.

In Fig. 8d it can be seen that the share of CH₄ in THC emissions has risen compared to RDE Moderate, which is in agreement with the results comparison of Tables 6 and 7. This is a consequence of higher loads and richer operating conditions compared to the RDE Moderate route resulting in a more intense formation of CH₄ from higher hydrocarbons during the RDE Extended route.

Similar to the CH₄ and THC emissions, it can be seen that the CO emissions (Fig. 8e) are much more pronounced at high load, i.e. around 1800 s when the vehicle began its ascent and between 3800s and 4700s when the vehicle was driven on the motorway. The conversion efficiency of CO in a TWC is considerably reduced due to the rich mixture operation [33], therefore, notable CO emissions in sections with high loads are expected. In contrast to CO emissions of the RDE Extended route when using CNG, the CO emissions behave in an expected manner, therefore, it can again be concluded that the measuring error seen in the CO emissions is not present when using gasoline. Additionally, a similar magnification inspection as conducted when analyzing the CO emissions of the RDE Moderate route when using gasoline was done. It revealed no irregularities nor indication of any CO emission measurement errors.

It can be seen that during the RDE Extended route NO_x emission (Fig. 8f) peaks occur more frequently indicating more sudden load increases and decreases compared to the RDE Moderate route when using gasoline. NO_x emission peaks are, in general, of similar ppm values compared to the NO_x emission peaks of the RDE Moderate route using gasoline. The NO_x emission peak at 2800s, when the vehicle was descending, is a consequence of a load increase at lean operating conditions leading to NO_x breakthrough due to low EG temperatures and excess oxygen.

3.4. Consistency of CO measurements

When conducting measuring campaigns using a PEMS, the attention is primarily focused on cumulative emissions; hence, any unusual behavior of the instantaneous time-dependent emission traces can often be overlooked. Although the cumulative CO emissions of both RDE-based routes when using CNG as fuel in the present analysis are well below Euro 5 emission limits, our measurement campaign reveals that a problem can occur during the measurement of CO emissions. Additionally, a similar trend of CO emissions was also present during the WLTC when using CNG as fuel and on other routes using CNG conducted on Day 4 of this measuring campaign. Figs. 4 and 7 indicate two types of CO emission peaks occurring during the test. The ones that show nonrealistic time dependency are characterized by a rapid rise followed by an exponential fall of the CO emissions. Furthermore, the nonrealistic CO emission peaks coincide with engine load decrease and thus very low fueling or a fuel cutoff. Under given conditions, such high levels of CO emissions are unlikely to occur, especially as they are not accompanied by an increase of THC emissions. As shown in Appendix C, these false positives can be with high certainty linked to the cross sensitivity of the measuring method of CO and N₂O, which could possibly be formed under these operating conditions.

3.4.1. N₂O formation in the TWC converter of an SI internal combustion engine when using CNG

Emissions of N₂O from vehicles are a well-recognized environmental issue, primarily on the account of N₂O being a potent greenhouse gas [38, 40, 41 and 42]. Several studies reveal that N₂O can be detected in catalyst-out emissions ranging from a few tens of ppm to near 800 ppm [35, 37, 39, 40 and 42]. In [38, 40], it is reported that formation of N₂O proceeds via reduction of NO by CO when O₂ is present. Similarly, the study in [49] reports that N₂O formation is favored under slightly lean operating conditions. The N₂O formation is observed primarily during light-off at fairly low temperatures ranging from 110-400°C [38, 40, 42 and 45]. In [39, 42 and 45], it is stated that N₂O decomposes at temperatures over 500°C. In [53], it is reported that under real operating conditions significant release of NO may take place and accordingly N₂O formation is likely to occur.

The TWC converters were originally designed to reduce engine-out emissions when using gasoline. The majority of modern TWC converters used in SI internal combustion engines thus rely on Pt, Rh and Pd [26] as active metals. However, converters that have been adapted for CNG vehicles mainly use Pd catalysts, which have the highest activity for CH₄ oxidation [26] and are suited to the high concentration of H₂O in the exhaust gasses and to the typically transient reaction conditions that result from cycling between oxidizing and reducing conditions when using CNG. These are known to significantly influence the catalyst's activity and stability [26]. Typical exhaust gasses when using CNG are characterized by low temperatures (even below 400°C) and higher amounts of H₂O vapor (due to the high content of hydrogen in methane) than gasoline use. In [46] it was reported that engines with a CNG injection system suffer from λ overshooting due to drifts of injected fuel mass caused by pressure drops in the system when adjusting to required loads. They reported that dedicated and refined control strategies capable of preventing mass mismatches, negatively affecting the aftertreatment system, are needed [46]. Furthermore, in [51] the existence of strong nonlinearities in the lower part of the gas injector flow chart compromising the air-fuel ratio was exposed. In [52] it is reported that due to the gaseous nature of the fuel, pressure waves are present within the fueling system. In addition, it is stated that the liquid fuel in a gasoline injector acts as a damper on the injector needle as it is opening and closing, whereas a CNG injector needle will bounce when opening and closing, causing pulsations of fuel to leak through the opening [52]. It was concluded that at high engine speeds these pulses could account for up to 1/3 of the total fuel injected [52].

The conditions where a combination of above-listed phenomena could occur are rapid changes in throttle position, particularly rapid cut-offs of fuel supply which follow sections with relatively high load demand. These conditions closely coincide with an exponential decrease of measured CO emissions, which were pinpointed as false positives. Considering the listed background on N₂O formation, specifics of the TWC converters and the CNG injection systems causing CH₄ leakage as well as higher amounts of H₂O vapor in the exhaust gas when using CNG, the most plausible explanation of N₂O presence in the exhaust gas sample fed to the PEMS during the measuring campaign is the N₂O formation in the TWC when the vehicle is using CNG. Since the false CO readings follow an exponential relation, they can be correlated to the Arrhenius equation [36] and the chemistry kinetics. To further confirm the relation to the chemistry kinetics, a short analysis of governing reactions for N₂O formation is presented in Appendix B.

3.4.2. Supporting evidence of the hypothesis through instantaneous RDE measurements

Fig. 9. Visualization of instantaneous on-road NO and CO emissions of RDE Moderate route using CNG

Fig. 10. Visualization of instantaneous on-road NO and CO emissions of RDE Moderate route using gasoline

Figs. 9 and 10 represent the visualization of instantaneous on-road NO and CO emissions of RDE Moderate when using CNG and RDE Moderate route when using gasoline, respectively. Fig. 9 clearly indicates a correlation between NO emission peaks and false CO emission peaks when using CNG on the RDE Moderate route. However, this correlation is non-existing in Fig. 10, which represents RDE Moderate route when using gasoline.

These results can thus be considered as supporting evidence of the hypothesis introduced in the previous parts of this section. In Fig. 9 it can namely be seen that almost every NO emission peak is accompanied with a λ peak, indicating a significant decrease in load. It is also seen in Fig. 9 that the unusual CO emission peaks are detected after a substantial rise of λ . It is hypothesized that the CO emission peak followed by an exponential decrease is, in fact, the NDIR measuring N_2O generated by (B.1), (B.2) and (B.7) and yielding false CO reading. In sections where NO drops to zero, it is speculated that N_2O formation ceases, hence the exponential decrease of CO is observed, indicating the decomposition of N_2O . There are several indicators, such as the correlation of the CO exponential falls to the Arrhenius equation [36] and to the chemistry kinetics laid down in Appendix B, proving that the CO false readings are not an aftermath of contamination of the analyzer with e.g. liquid water which would produce false CO and CO_2 readings that would not follow any given chemistry relations.

4. Conclusion

In this paper, an RDE-based assessment of a Euro 5 factory bi-fuel CNG/gasoline light-duty vehicle equipped with the TWC was presented. The vehicle specifications were first evaluated considering the WLTP procedure followed by an appropriate selection of the WLTC cycles. Considering the RDE legislation trip boundary conditions two RDE-based routes were derived: an RDE Moderate route, satisfying the normal conditions and representing a conventional route, and an RDE Extended route, satisfying the extended conditions and representing a still plausible real-life driving route.

The test vehicle's main fuel is CNG, therefore the engine has a higher compression ratio than a conventional spark ignition gasoline engine, which means that the gasoline mode in this bi-fuel engine does not completely mimic the behavior of a typical gasoline engine. In order to avoid knocking, the manufacturer had to optimize the ECU maps when the vehicle enters the gasoline mode. Nevertheless, the emissions of such vehicles in gasoline mode are not to be neglected, since the rarity of CNG fuelling stations and on the other hand, the frequency of gasoline pumps often forces such vehicles to run on gasoline mode. It is also important to note that the equipped TWC was designed to adequately perform on an NEDC cycle. Therefore, it was expected that some discrepancies from Euro 5 limits would be present when driving a more intensive and power-demanding route due to the breakthrough of the emissions resulting from insufficient capacity/sizing of the TWC.

The original contributions of this paper comprises of analyses of the RDE-LDV emissions of a CNG-powered vehicle, benchmarking of the CNG RDE-LDV results with the ones measured for gasoline in the same vehicle and under similar operating conditions, exposing the strong interference of CO and N₂O infrared absorption bands, which can significantly affect the CO readings from the non-dispersive infrared sensor (NDIR), revealing the hypothesis why CO and N₂O interference is more pronounced when using CNG in LDVs and reasoning of the time resolved on-road real driving emissions trends. For instance, the sudden load decreases and increases throughout the RDE-based routes accounted for most of the NO_x emissions. The uphill driving section when using gasoline proved to emit the bulk of the cumulative THC, CH₄ and CO emissions, whereas the downhill driving section when using CNG accounted for most of the THC and CH₄ emissions. These effects are much more pronounced in the RDE Extended route. The in-depth analysis of the CO emissions when using CNG indicates that a significant decrease in load can be correlated with a CO measuring error.

This study has highlighted the difficulties of measuring CO emissions of bi-fuel CNG/gasoline vehicles when using CNG. CO emissions of both RDE-based routes when using CNG indicate that the NDIR analyzer produces a CO emission measurement error since this phenomenon is seen on both RDE-based routes. The detection of this phenomena was made possible by analyzing the multitude of new driving patterns, such as the downhill sections, the uphill sections, several full stops and accelerations in the urban sections, the presence of traffic, driver behavior, etc., which are significantly different to those that can be encountered in the NEDC. The measuring campaign test sequence indicates that the measurements using CNG were not conducted successively nor on the same day and that the RDE Extended route was not interleaved with the RDE Moderate route as they were conducted on separate days, therefore, the possibility of a single point in time instrument failure can be with high certainty excluded. In an attempt to additionally investigate this phenomenon, an experiment was conducted proving the influence of N₂O presence in the gas fed to the PEMS on the CO readings of the NDIR analyzer. Experimental results indicate that even the slightest traces of N₂O in the gas fed to the PEMS can lead to severe CO misreading. Specifically, the experimental results suggest that values between 100 and 600 ppm could induce such anomalous CO signals. The

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suggested estimate is in line with the literature review, which suggests that N₂O can be detected in catalyst-out emissions ranging from a few tens of ppm to near 800 ppm. Supporting this claim, several hypotheses of N₂O formation were also presented in the paper. Furthermore, the paper also presents reasoning, based on the entire route from the mixture preparation over engine-out species composition and temperatures to chemical reactions in the TWC, why these interference issues are more significantly pronounced while using CNG. This study thus opens challenges related to CO emissions of NDIR analyzers that go far beyond the presented analysis. The revealed interference between CO and N₂O measurements, causing false positives of CO is strongly linked to the methodology of the measurement. Devices, based on other than NDIR method for CO/CO₂ measurement are less sensitive to this issue, however, most of the market available PEMS are equally affected, as they strongly rely on NDIR for CO/CO₂. In this case, the reliability of CO measurement with CNG is similar among different manufacturers.

The highlighted time resolved on-road real driving emissions trends resulted in the cumulative emissions, which were compared against the Euro 5 regulatory limit. The indicative comparison reveals that the emissions of THC, NMHC, and NO_x of both RDE-based routes using both fuels were under the Euro 5 limits. Additionally, the THC and NO_x emissions of both RDE-based routes using both fuels were under the type-approval limits. A comparison of the NO_x emissions of both RDE-based routes using both fuels reveals that the vehicle emitted more NO_x when using CNG, which is in line with other studies and is also reasoned with the time engine parameters and time traces of the emissions. The CO₂ emissions of both RDE-based routes when using both fuels were above the type-approval limits with CNG exceeding the type-approval limits by 6-35% and with gasoline exceeding the type-approval limits by 66-85%. The comparison of both RDE-based routes also reveals that the CO₂ emissions were roughly 30% lower when using CNG, which is in line with the literature review. Since a CO emission measurement error is detected on both RDE-based routes when using CNG, an assessment of the CO emissions when using CNG is unreliable for this fuel. The CO emissions of both RDE-based routes using gasoline were above the type-approval and Euro 5 limits with the CO emissions on the RDE Moderate route exceeding the type-approval limits by 920% and the Euro 5 limits by 190%. The CO emissions of the RDE Extended route when using gasoline exceeded the type-approval limits by 1160% and the Euro 5 limits by 242%.

APPENDIX A. WLTP and RDE Legislation background and procedure

The WLTP consists of four phases with different velocity distributions (low, medium, high and extra high). Based on the vehicle's power-to-mass ratio and its maximum velocity three different driving cycles were developed representing three different vehicle classes [6, 7]. The length of the cycle is fixed at 1800 seconds. Gear shift points are determined mathematically and are based on the characteristics of individual vehicles [7]. In addition to the driving cycle, a procedure is also introduced to prescribe test conditions, requirements, tolerances, etc. General requirements of the vehicle state that the manufacturer shall specify the types and amounts of lubricants and coolant, as well as the tire specifications for emission testing, intended for normal vehicle operation [7]. Testing shall be performed considering several Annexes, which are in detail explained in the Global Technical Regulation no. 15 (GTR.15) [7].

Complementary to the WLTC tests, the RDE test is intended to be introduced. This test is performed in addition to the laboratory tests and is designed to check emissions outside of well-established testing procedures in laboratories. In practice, RDE implies testing of new vehicles on the road using Portable Emissions Measurement Systems (PEMS). The general

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requirements and conditions of the RDE test are formulated and explained in the Appendices of Annex IIIA [8]. During the RDE test, the vehicle should not emit higher emissions than the not-to-exceed (NTE) values:

$$NTE_{pollutant} = CF_{pollutant} \cdot EURO\ 6, \quad (A.1)$$

wherein the currently proposed 2nd package of the RDE legislation [27] the value of $CF_{pollutant}$ (conformity factor of the pollutant) for NO_x is from 09/2017 equal to 2.1 and the final value from 01/2020 is 1 + a margin of measurement uncertainties. The margin for measurement uncertainties is subjected to annual reviews and shall be revised because of the improved quality of the PEMS procedure or technical progress [27].

APPENDIX B. Governing equations supporting the N₂O formation in the TWC when using CNG

Several chemical reactions are proven to produce N₂O over supported rhodium, platinum and palladium catalysts [40]. The following reactions are enhanced by the presence of O₂ [40]:

Additional H₂ and CO for reactions in eqns. (B.1) and (B.2) can be produced via water-gas shift reaction

and methane steam reforming:

which are more pronounced when using CNG due to a higher concentration of H₂O vapor compared to gasoline use. The CH₄ required for (B.4) and (B.5) can be associated with the previously listed specifics of the CNG injection systems causing CH₄ leakage.

An additional N₂O production route in a TWC (B.7) has been suggested in [42]. NH₃ is formed in the 200-600°C range via reduction of NO by H₂ (B.6) [38], which is produced from reactions in eqns. (B.3), (B.4) and (B.5). Figs. 4, 7, B3 and B4 reveal that each Lambda increase is followed by a nonrealistic CO emissions peak. It can thus be hypothesized that the presence of NH₃ in combination with an abundance of O₂ caused by said Lambda increases could subsequently lead to N₂O formation via (B.7):

To fully confirm the hypothesis of N₂O presence in the exhaust gas having an impact on the CO readings of the NDIR analyzer, an experimental evaluation of the cross sensitivity of analyzers is provided in Appendix C.

APPENDIX C. Cross sensitivity of CO and N₂O in an NDIR analyzer

The PEMS used in this measuring campaign was equipped with a non-dispersive infrared (NDIR) sensor for CO and CO₂ measurements, which is a widely accepted method among PEMS manufacturers. NDIR gas detection method is based on the absorption of specific wavelengths of infrared radiation by different gaseous species. Ideally, species that are relevant for analysis should exhibit distinctively different footprints in the infrared spectrum resulting in minimum overlap of the absorption bands. Where overlapping is expected, the cross sensitivity is respected in calibration procedures, reducing the impact on measurement accuracy. Based on the results of this study, strong indices were identified that N₂O emissions could significantly influence the PEMS measurements accuracy of CO emissions through this mechanism.

The interference was already confirmed in the case of FTIR analyzers [50], which use more complex methods of analysis (detailed spectral analysis instead of only band-pass analysis, typical for an NDIR analyzer). It is reported that the measurements of N₂O concentrations in diluted exhaust samples and on-line real-time analyses were disrupted by interference from CO absorption bands [50]. In [41], it is reported that NDIR analyzers are even more affected by the presence of interference gasses than FTIR analyzers, primarily due to the lack of selective optic and detecting systems.

Fig. C1. Infrared spectrum of carbon monoxide (CO) and nitrous oxide (N₂O) [34]

Fig. C1 reveals that the infrared spectrums of CO and N₂O are similar, especially in the interval of highest absorbance, which could induce CO emission measuring errors when using NDIR if the analyzed gas contains traces of N₂O. Therefore, in the context of this study, an experiment with a similar commercially available analyzer of another manufacturer was performed to confirm the influence of N₂O presence in the gas fed to the NDIR-equipped PEMS on measured CO emissions as observed in on-road tests. With this objective in mind, the bag method was adopted. First, an exact volume of gas with a known composition (synthetic air and calibration gas) was fed into the binning bag. Secondly, an adequate amount of N₂O was introduced into the bag to reach the desired N₂O concentration. The gas from the bag was then fed to the PEMS. This procedure was repeated to obtain the following concentrations of N₂O in the feed gas: 0 ppm, 100 ppm, 200 ppm, 400 ppm and 600 ppm.

Results in Fig. C2 illustrate the effect of N₂O presence at various concentrations in the synthetic air with 0 % CO and in calibration gas with 0.4262 % CO on the outcome of CO emission measurements. It can be seen that the presence of N₂O in the analyzed gas has a considerable effect on the CO emission measurements in an NDIR cell. The relation between N₂O ppm content in the analyzed gas and measured CO emissions is almost linear with both carrier gasses (synthetic air and calibration) in both gas variations. These results further strengthen the hypothesis that the presence of N₂O in the analyzed gas fed to the NDIR-equipped PEMS causes false positives of CO measurements.

Fig. C2. Effect of N₂O presence in various mixing ratios in synthetic air with 0% CO (green markers) and in calibration gas with 0.4262% CO (red markers) on PEMS-measured CO emissions

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