Particle emissions measurements on CNG vehicle focusing on, sub-23nm

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Abstract

Current study aims to investigate sub-23nm solid particles (SPN<23nm) from spark ignition engine using compressed natural gas (CNG) as a primary fuel working in port fuel injection (PFI) mode and gasoline working as direct injection (GDI). For that reason, a state-of-the-art exhaust gas sampling system used for solid particle detection up to 2.5nm. Vehicle tested in various driving cycles. The SPN<23nm investigation have shown particle emissions beyond EURO 6 limits for both fuels. During tests, Particle Size Distribution (PSD) profiles reveal nucleation mode characteristics for both fuels. Though geometric mean diameter (GMD) for CNG operation was smaller, indicating that different combusting phenomena can formulate smaller in size particles.

Introduction

CNG market penetration increased over the years, reaching on average 6% annual increase with over 1.1 million vehicles running with natural gas in Europe (NGVA Europe, 2017) (Transport & Environment, October 2018), with future trends to be more optimistic since natural gas vehicle registrations for 2018 doubled from previous year (ACEA, October 2018). CNG yields lower CO, CO_2 in comparison to gasoline and lower CO, CO_2 and NOx emissions in comparison to diesel (Owen, et al., 1995) (Yusaf, et al., 2010) (Jahirul, et al., 2010) (Cachón, et al., 2009). Lower price as well as lower gaseous emission impact, are the main drivers for increasing market share of natural gas.

Gaseous emissions of CNG equipped passenger cars have been thoroughly investigated over the years, but for particle emissions perspective research is limited. Generally, exhaust tailpipe particle emissions below 100nm are harmful to public health as they can easily penetrate via breathing into the pulmonary system (Stone, 2012) (Eastwood, 2008) (Hinds, 1982). Research on sub-23nm particle emission in CNG passenger cars is limited. To this moment, studies on CNG particle emissions, focus on retrofitted engines (Kento T. Magara-Gomez, 2014), heavy-duty vehicles (Xue, et al., 2018) (Nylund, et al., 2004) or CNG-Diesel and CNG-Gasoline combined operation, with parallel investigation of engine operation characteristics (Jahirul, et al., 2010) (Yusaf, et al., 2010) (Nithyanandan, et al., 2016). In one case, experimental optical engine was used to conduct particle emission study performance in dual fuel operation with various blends including CNG in direct injection mode (Catapano, et al., 2017). Recent and most relevant research in the field investigated the potential difference between CVS and tailpipe particle emissions measurements including CNG equipped passenger cars (Giechaskiel, et al., 2019). In general, the previous results have shown a general trend of higher particle emissions towards nucleation mode region in CNG operation as compared to Gasoline operation.

As regards testing methodologies of previously mentioned studies, those, were restricted to steady state points or outdated driving cycles (NEDC) (Catapano, et al., 2017) (Mayer, et al., 2012) using indirect methods of sampling (CVS) (Schreiber, et al., 2007) in most of the cases. Indirect method of sampling poses a potential risk of lab to lab high measurement variability (Mamakos, et al., 2004) (Mathis, et al., 2004) and can lead to misunderstanding of actual PSD and thus to different aggregated particle emissions from tailpipe to CVS (Cedric, et al., 2017) (Giechaskiel, et al., 2019). Current work addresses these issues by implementing direct tailpipe sampling technology, which minimize the potential risk of particle losses. This work aims to investigating sub-23nm particle emissions using direct tailpipe measurement. For that reason, the DownToTen (DTT) sampling system was developed and funded under the framework of the European Commission's HORIZON 2020 program (Samaras, et al.) (Loctier, 2019).

Methodology

Current study is focusing on sub-23nm particle emissions investigation on a CNG equipped vehicle. To accomplish this, a bi-fuel passenger car investigated. The vehicle has CNG as the primary fuel and Gasoline as a secondary fuel that used when CNG is depleted. Fuel injection technology differs from CNG to Gasoline, the primary fuel injection is PFI and the secondary, is GDI technology. Gasoline interinjection, when the engine is running in CNG, is not required since the combustion chamber designed to disperse excessive heat (Markowski, 2017). To have a holistic view of particle emissions, a combined measurement campaign was adopted using both fuels in series, during experiments. Testing protocol included series of driving cycles in the chassis dyno first with CNG and then with Gasoline fuel. Engine characteristics are presented below (Table 1).

Table 1 Test vehicle characteristics

Vehicle segment	C
Engine	Spark-ignition 4-cyl, 16-valve
Model Year	2018
Fuel	CNG (PFI) and Gasoline (GDI)
Drive, Transmission and Number of gears	FWD, Manual, 6
Maximum power [kW] / Torqure [Nm]	81 at 4800-6000rpm / 200 at 1500-3500rpm
Engine capacity [cm^3]	1395
Engine Stop-Start system	Yes
Emission Standard	Euro 6b
Aftertreatment system	TWC

SPN<23nm particle detection acquired using DTT exhaust gas sampling system. The system consists of two porous tube dilutors, the first dilutor is heated to avoid condensations, along with a catalytic stripper (CS) and ejector diluter (ED). In the DTT sampling system three particle detection devices attached. Two of them were CPC based and the other one using particle charging technology (EEPS). The DTT system addresses the issues that previous researchers raised regarding the sub-23nm measurement feasibility (Giechaskiel, et al., 2014). Thus, DTT system yields a dilution system with low particle losses in the sub-23nm region with a CS optimized for accurate determination of sub-23nm particles. In particular, by introducing porous tube dilutor, thermophoretic losses were reduced to almost zero. The dominating source of diffusional losses are reduced by optimizing – downsizing the CS. Artefact formation was very low proving excellent performance.

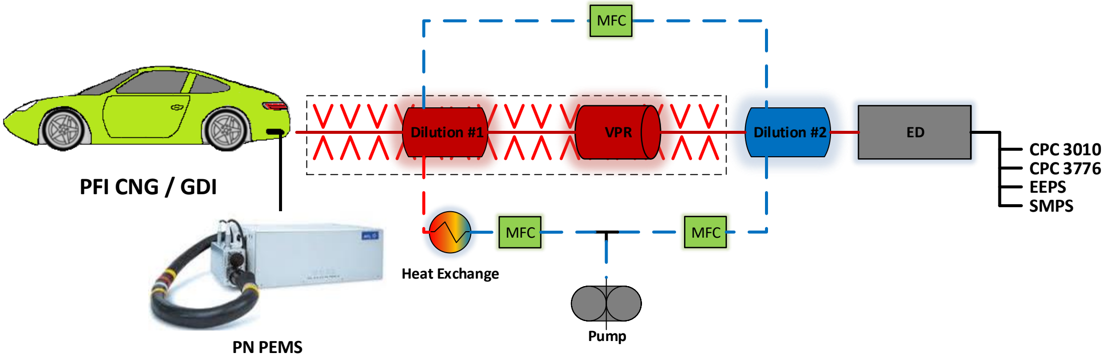

Figure 1 E.U. HORIZON 2020 DownToTen sampling system attached to the exhaust tailpipe. PN PEMS connected to the tailpipe for SPN>23nm measurements.

In addition to the DTT system that was sampling in the exhaust line, a PN PEMS device was installed to the tailpipe for SPN>23nm measurements. The PN PEMS is a PMP compliant device, based on diffusion charging technology. During steady state runs an SMPS classifier also used to acquire particle size distributions. In order to avoid artefacts during measurements, the exhaust line was covered with insulation. Technical specifications of the measurement devices are included in the following table (Table 2).

Table 2 Technical characteristics of particle detection instrumentation

	AVL PN PEMS	***SMPS, TSI 3080***	***CPC, TSI 3776***	***CPC, TSI 3010***	***EEPS, TSI 3090***
Particle size (nm)	SPN>23 (PMP)	10 - 1000	2.5	10	5.6 - 560
Concentration range (#/cm^3)	3000 – $2*10^7$	108	0 – 3*105	0 - 10000	300 – 107 (5.6nm) 3 – 105 (560nm)
Time resolution	10Hz	30-120s	1Hz	1Hz	10 Size distributions/s
Particle detection technology	Diffusion Charging	Neutralizer and CPC	CPC based	CPC based	Unipolar charging

The Bi-fuel passenger car considered for this study was tested on steady state runs as well as under several driving cycles. Tests performed on a one-axis chassis dynamometer using road load settings calculated from a coast down test. Three types of test cycles were used, including WLTC, NEDC and SRC. Main idea was to conduct measurements in various test cycles to have a better understanding of particle emission performance under different engine loads.

In order to acquire a large amount of experimental data, 23 laboratory tests were performed on the chassis dynamometer. Engine preconditioning and soaking was mandatory prior to each measurement day. Warm up time, calibration and zeroing it was applied to every device so as to maintain high level of measurement validity. Prior the chassis dyno testing, a coast down test was performed to acquire the real-world road load. Regarding fuel testing sequence, CNG fuel tested at first since it was the primary fuel since no selection capability between two fuels was applicable. After depletion of CNG, engine ECU switched automatically to Gasoline. Typical fuel properties of CNG and gasoline are listed in Table 3 as extracted from literature (Heywood, 1988) (Vogler, et al., 2018).

Table 3 Fuel characteristics

Parameter	**Gasoline E0**	**CNG**
Chemical formulae	C4-C12	CH4
State at NTP conditions	Liquid	Gas
Density at STP [kg/m^3]	730	0.79
Octane number [-]	84-95	120+
Lower Heating Value [MJ/kg]	43.5	48.5
H/C ratio [-]	2.0:1	3.9:1

Results and discussion

In Figure 2, the aggregated particle emissions [#/km] for both fuels are depicted. With light green background colour are the CNG results whereas with light yellow background colour are the Gasoline ones. As it can seen, blue bars which correspond to SPN>23nm particle emissions measured with PMP compliant PN PEMS, are all within current EURO 6 limits ($6*10^{11}$ #/km). As the particle emission investigation expands to a greater range, emission results are increasing for both fuels. Emissions results for SPN>10nm, SPN>5.6nm and SPN>2.5nm, are in the same level for both fuels. This indicates that PFI CNG operation reveals comparable particle emissions with GDI mode in the sub-23nm region. In addition to that, sub-23nm particle emissions are beyond EURO 6 limits in almost all particle size ranges.

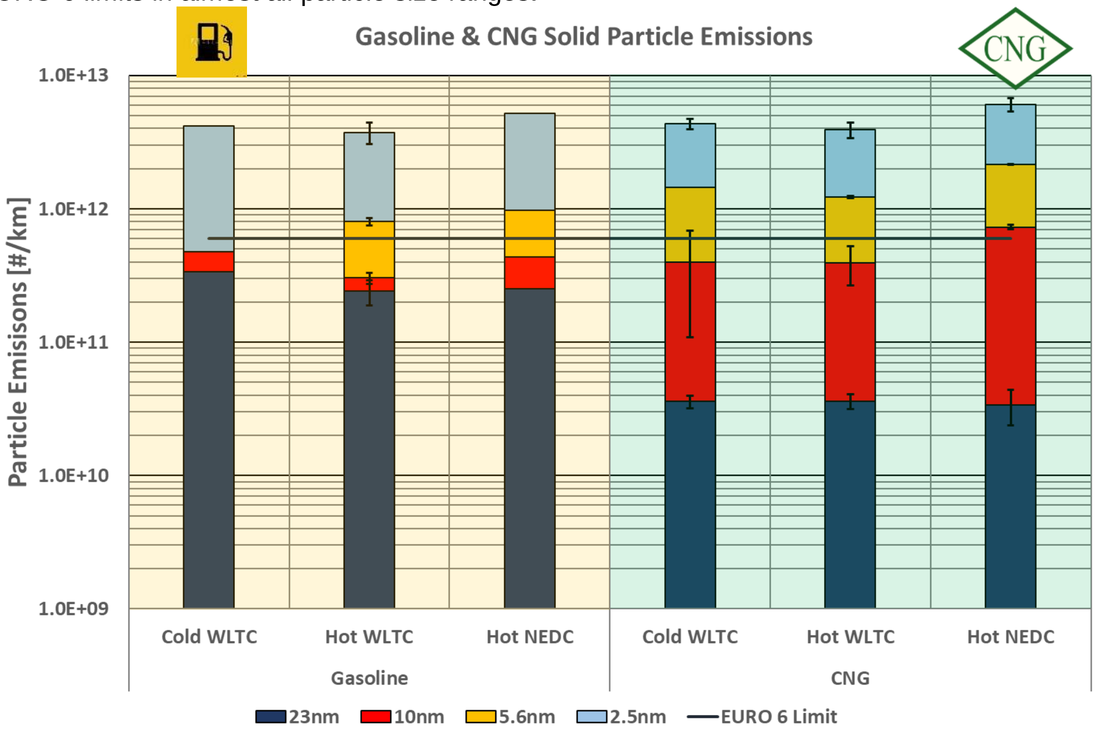

Figure 2 Aggregated SPN emissions for Gasoline (light yellow) and CNG (light green). Error bars shown the standard error of the mean including 2 to 4 repetitions. Emission bars with no error bar indicating unique measurement. Emission limit corresponds to EURO 6 ($6*10^{11}$#/km)

Figure 3 illustrates the SPN ratios together with mean particle concentration values of SPN>23nm. In particular, SPN10nm/SPN23nm, SPN5.6nm/SPN23nm and SPN2.5nm/SPN23nm are presented from darker to lighter green. As can be seen, CNG SPN10nm/SPN23nm ratios are much higher compared to Gasoline values. This is indicative of higher sub-23nm particle emissions during CNG than Gasoline operation. In other words, GMD of CNG is shifted towards nucleation mode particles.

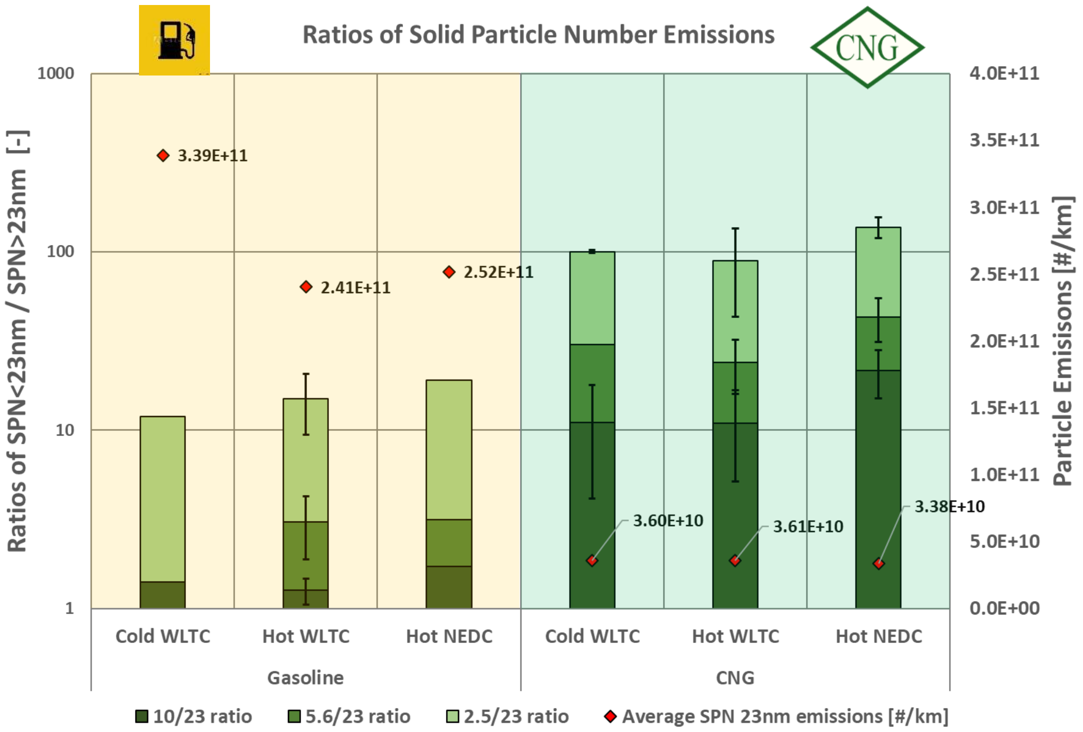

Figure 3 SPN ratios bars are depicted form darker to lighter green. Average values of SPN>23nm particle emissions are presented with red diamonds. Error bars shown the standard error of 2 to 4 repetitions. Emission bars with no error bar indicating unique measurement.

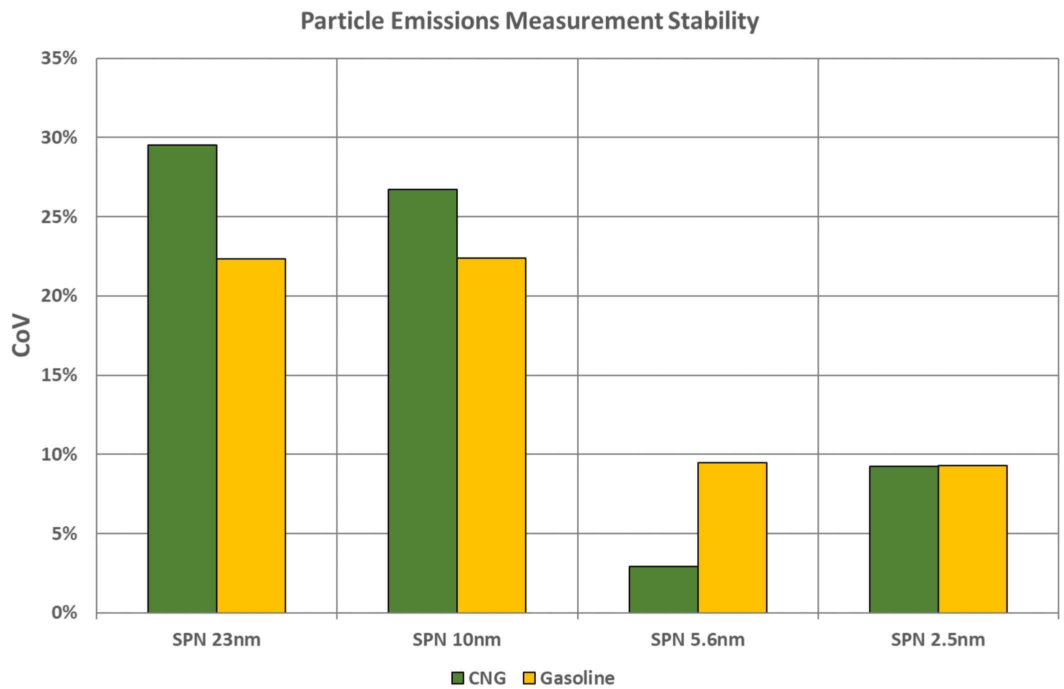

Figure 4 Particle emission measurement stability for each cut off size category.

In Figure 4 the measurement repeatability is depicted. For that reason, the statistical parameter of covariance (CoV) was used, which is the standard deviation of each measurement category over the average value of all measurements in the same category. There are four categories that correspond to different cut-off particle sizes. CoV function is presented herein.

$$CoV_{SPN} = \frac{S_{SPN}}{\overline{SPN}}$$

Figure 4 results, after outlier removal, shown significant drop from 30% for CNG and 22% for Gasoline to 9% for both fuels. Since this engine produces particles in the ultrafine region below 23nm, as the measurement widens to sub-23nm particle detection, measurement reproducibility is getting optimized since the nucleation mode captured better from the sub-23nm particle detection devices. Same trend also revealed from other researchers that were investigating in the sub-23nm region (Giechaskiel, et al., 2019). CoV of Gasoline has better reproducibility than CNG

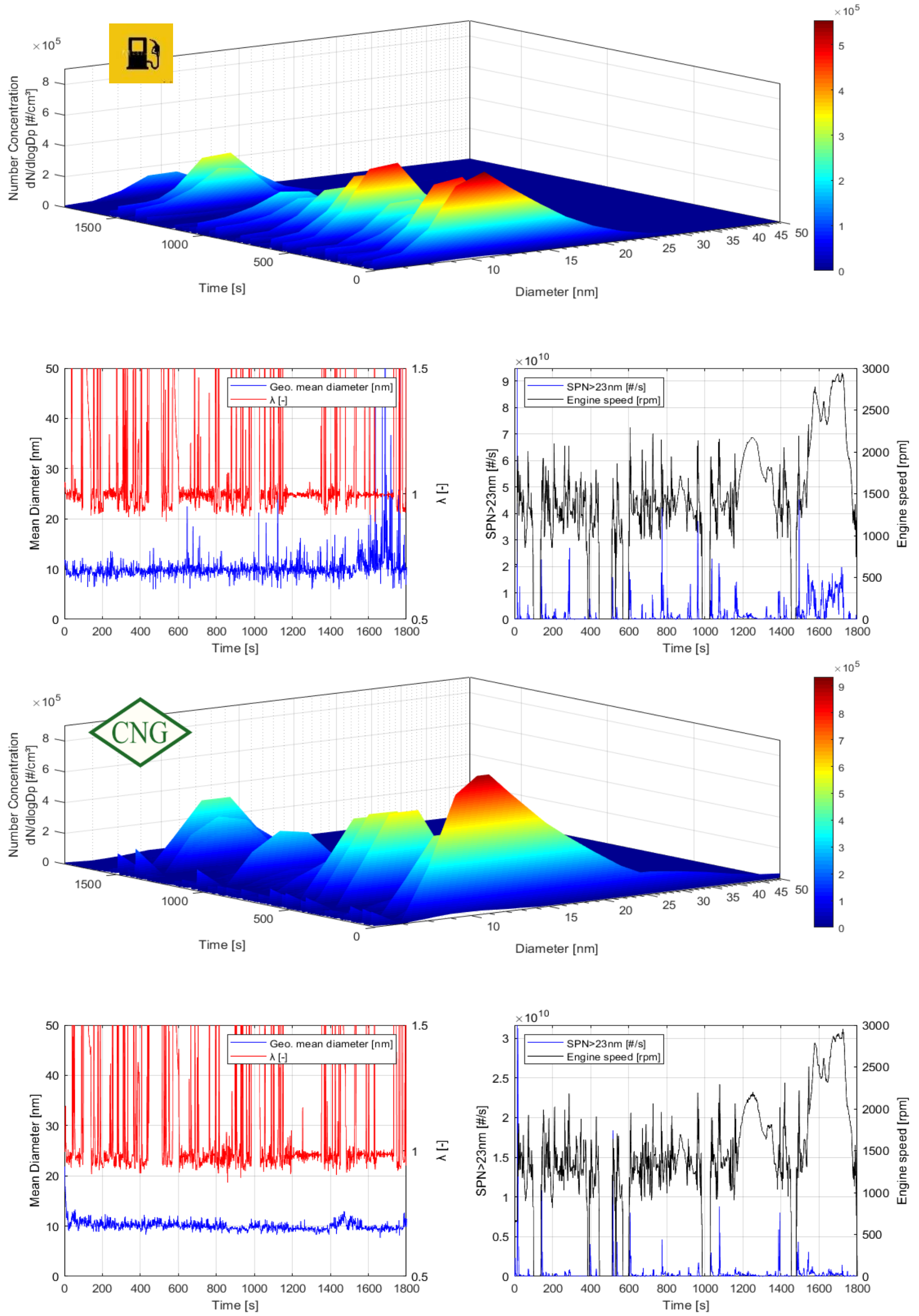

Figure 5 PSD profiles (3D map), with Lamda (red, bottom left) Dg (blue, bottom left) and SPN>23nm particle emissions (blue, bottom right) for Gasoline (top) and CNG (bottom) during Hot WLTC

from early stages, since Gasoline PSD is shifted towards a greater GMD and thus, particle detection with reproducible manner from early stages. CoV parameter for both fuels was constantly below 10% after SPN<5.6nm.

In Figure 5 the time series of particle concertation, GMD and PSD profiles are presented. Figure data referred to Hot WLTC measurements for Gasoline (top) and CNG (bottom). GMD of gasoline is higher than CNG, as it can be seen at the bottom left graph, that confirms initial finding of CNG smaller particles. What is remarkable is that GMD of GDI tends to increase in higher loads in contrast to CNG PFI which is getting slightly lower during high loads. In addition, large peaks of GMT being observed under heavy loads, immediately after fuel cut events, during fuel enrichments (equivalence ratio in red) that is leading to the fact that fuel impingement is prevalent when engine is running under GDI mode. In CNG PFI operation same events cannot be seen.

Those two trends indicate that the PSDs of GDI and PFI CNG change during the cycle and strongly depended to the engine load. As regards GDI GMD behaviour, this, mainly has to do with higher carbon content of gasoline which corresponds to soot related particles during rich mode and high engine loads. Wall impingement of fuel spray should not be neglected, as it is largely affecting pool fire events and thus soot related particles. Regarding GDI engine particle emission performance there is ample piece of information that reader may find in the literature (Raza, et al., 2018).

On the other hand, the absence of C-C bonds in CNG PFI and the low carbon content (Table 3) aren't able to promote particles in larger diameters. One possible explanation of nucleation mode particle emissions during CNG PFI operation is the lubrication. Lubrication related particle emissions can be attributed to escaping oil from valve stems and piston rings which could be a sign of engine wear. Another source is the exposed bore oil during combustion which could be evaporate in the exhaust line (Eastwood, 2008), that potentially generate nucleated particles under intense combustion process (Mayer, et al., 2010). Since no significant particle emissions can be observed during deceleration phases (Figure 6 deceleration phase within red dashed circle), which can be attributed to escaping lube oil, high sub-23nm particle emissions can be attributed to exposed lube oil during combustion. The combination of different CNG fuel characteristics when compared to gasoline and the exposed bore oil under different combusting phenomena that occurring in CNG (Vogler, et al., 2018) (Catapano, et al., 2017), is able to magnify sub-23nm particle emissions.

Instantaneous particle emissions measurements are presented in Figure 6. Sub 23nnm particle emissions including SPN>2.5nm (orange) and SPN>10nm (red) are also depicted. In red dashed circle the final phase of WLTC cycle (extra high) is underlined for both fuels. What can be seen here is that PFI CNG operation reveals higher SPN>2.5nm emissions than GDI engine operation. The extra high WLTC phase corresponds to higher engine loads, thus, it can be concluded that CNG PFI engine operation emits higher sub-23nm particle emissions than GDI during high engine demand.

As regards ignition timing, illustrated in blue dots, the studies that includes both particle emissions measurement and ignition timing evaluation are limited. The majority of these studies are focused on GDI engines without GPF, along with or without TWC under steady state operating points instead of transient ones, such as the current case (Ketterer, et al., 2014) (Price, et al., 2007) (Qin, et al., 2014). The general trend, as previous researchers discovered, is that as the ignition timing advances, particle emissions increase during steady state operation. Having in mind the measurements conducted under transient conditions during this study, no clear evidence of strong correlation between ignition timing and particle emissions can be seen until the three quarters of the cycle. Advanced ignition timing promotes higher exhaust gas temperature inside the cylinder, which can also contribute to nucleation mode. This trend also revealed during CNG PFI operation. From Figure 5, in the 1200s – 1600s region, as the ignition timing advances and towards higher exhaust gas temperature, engine load and speed, GMD decreases. Similar trend cannot be observed in GDI, in fact it's the opposite behavior that may indicate fuel impingement in cylinder walls that promotes soot related particles.

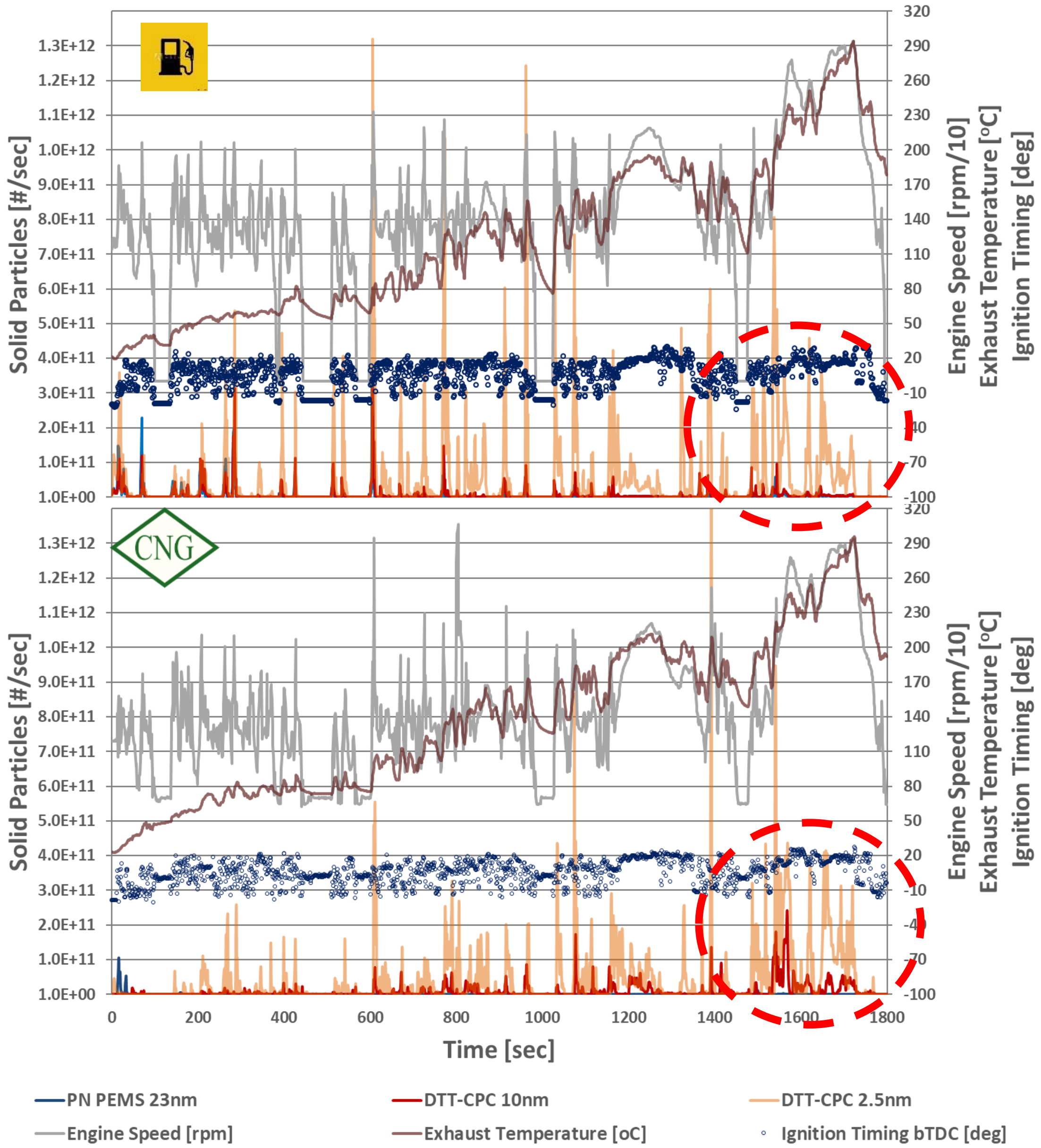

Figure 6 Instantaneous particle emissions including sub 23nm cut of sizes. The blue dots represent ignition timing events given in degrees of bTDC. The negative values represent retard ignition while the positive ones represent advance ignition. Exhaust gas temperature (pale red) measured at the sampling point. In the red dashed circle, the higher sub 23nm particle emissions of CNG can be seen when compared to Gasoline.

Summary and Conclusions

The purpose of this study was to investigate a CNG equipped passenger car in terms of sub-23nm particle emissions. Measurements performed using E.U. Horizon 2020 DownToTen sampling system that allows tailpipe testing using different particle detection devices. Conclusion of measurement campaign listed herein.

- CNG PFI engine operation reveals beyond EURO 6 limits particle emissions in the sub-23nm area. During measurements at the sub-23nm particle detection level, emissions were comparable to GDI engine operation.
- During CNG combustion, GMD was smaller compared to GDI operation, indicating that CNG PSD were located closer to nucleation mode particles.
- During high load GDI engine operation, the GMD was getting larger in contrast to PFI CNG operation which was getting smaller.
- During WLTC cold measurements, CNG operation has revealed higher sub-23nm SPN emissions in the extra high phase of WLTC in contrast to gasoline operation, this corresponds to higher sub-23nm SPN emissions than gasoline at higher engine loads.
- Sub-23nm particle emissions can be attributed to exposed lube oil during combustion. Further chemical characterization of sub-23nm particle emissions is needed.
- GPF installation considered as mandatory if the current regulation EURO 6 limit for sub-23nm particle emission is to be met.

Abbreviations

PSD: Particle size distribution
GMD: Geometric mean diameter
PFI: Port Fuel Injection
GDI: Gasoline Direct Injection
SPN: Solid Particle Number
CNG: Compressed Natural Gas
EEPS: Engine Exhaust Particle Sizer
CPC: Condensation Particle Counter
GPF: Gasoline particulate filter
TWC: Three-way catalyst
PEMS: Portable emission measurement system
ECU: Electronic control unit
CVS: Constant Volume Sampling
ED: Ejector diluter
CS: Catalytic stripper
WLTC: Worldwide harmonized Light vehicles Test Cycles
NEDC: New European driving cycle
SRC: Standard Road Cycle
PMP: Particle measurement programme

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Conflict of Interest

The authors declare that there is not any conflict of interest with this work.

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