Regulating particle number measurements from the tailpipe of light-duty vehicles: The next step?

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A R T I C L E   I N F O

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A B S T R A C T

Light-duty vehicle emission regulation in the European Union requires the dilution of the whole exhaust in a dilution tunnel with constant volume sampling prior to emission measurements. This methodology avoids measurement uncertainties associated with direct raw exhaust emission measurements from the tailpipe, such as exhaust flow determination, exhaust flow pressure pulsations, differences in the response time between exhaust flow and instrument signals, or their misalignment. Transfer tubes connecting the tailpipe to the dilution tunnel of different lengths, and mixing of the exhaust gas with the dilution air in the dilution tunnel may increase differences in measurements performed at different facilities. Recently, the light-duty vehicle regulation was complemented by on-road measurements with Portable Emissions Measurement Systems (PEMS). PEMS measurements are conducted from the vehicle tailpipe. Differences between tailpipe and full dilution tunnel measurements have not been adequately addressed so far. In this study we compare particle number emissions measured at the full dilution tunnel or directly at the tailpipe. The measurements covered solid particles with diameter larger than 23 nm, as required by the current regulation, but also solid particles larger than 10 nm, as recommended for future regulations. The studied vehicle technologies were diesel, gasoline, and compressed natural gas. The differences between tailpipe and dilution tunnel particle number emissions were found to be small (<15%) for both size ranges, with the exception of engine cold start (up to 35% in some cases). Theoretical estimates showed that agglomeration in the transfer line from the vehicle to the dilution tunnel might reduce particle concentrations by up to 17%. Exhaust flow rate determination and time misalignment of exhaust flow and particle concentration signals can introduce uncertainties of ±10% and ±5%, respectively, to the tailpipe measurements. The results suggest that tailpipe sampling is not only possible, but it can additionally give more representative (“real”) emissions of the vehicle and should be considered in post Euro 6 regulations.

1. Introduction

Air pollution is an important global risk factor for diseases (HEI, 2017). Ultrafine particles (smaller than 0.1 µm) have been associated with short-term cardiorespiratory and central nervous system adverse health effects (WHO, 2013). Traffic contribution to urban ambient particulate matter levels is estimated to be 5–80% (Pant and Harrison, 2013), but it can reach 90% in busy roads or even 99% in tunnels (Kumar et al., 2014). In the European Union (EU), the Solid Particle Number (SPN) of diameter >23 nm emission levels of diesel-light-duty vehicles have been regulated since 2011 (Euro 5b), and of Gasoline Direct Injection (GDI) light-duty vehicles since 2014 (Euro 6). According to the regulation, measurements are conducted after the exhaust gas is diluted with filtered air in a dilution tunnel with Constant Volume Sampling (CVS) (Giechaskiel et al., 2014b). The methodology is based on the recommendations of the Particle Measurement Programme (PMP) group (Giechaskiel et al., 2012b). It requires a primary hot dilution diluter at 150 °C, followed by an evaporation tube at 350 °C to remove semi-volatiles, and then a Condensation Particle Counter (CPC) with 50% detection efficiency at 23 nm (d50% = 23 nm) to count

Abbreviations: APC, AVL Particle Counter; CEN, Comité Européen de Normalisation; CNG, Compressed Natural Gas; CPC, Condensation Particle Counter; CS, Catalytic Stripper; CVS, Constant Volume Sampling; DPF, Diesel Particulate Filter; EEPS, Engine Exhaust Particle Sizer; EFM, Exhaust Flow Meter; ET, Evaporation Tube; EU, European Union; GDI, Gasoline Direct Injection; GMD, Geometric Mean Diameter; GPF, Gasoline Particulate Filter; JRC, Joint Research Centre; PCRF, Particle number Concentration Reduction Factor; PEMS, Portable Emissions Measurement System; PFI, Port Fuel Injection; PMP, Particle Measurement Programme; RDE, Real-Driving Emissions; SPN, Solid Particle Number; TP, Tailpipe; VELA, Vehicle Emissions Laboratory; VPR, Volatile Particle Remover; WLTC, World Harmonised Light-duty vehicles Test Cycle

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solid particles. This lower size (23 nm) was selected to include the typical smallest soot particles and to exclude semi-volatile nucleation-mode particles possibly surviving the evaporation tube. However, there are concerns that this lower size might not be appropriate for some vehicle technologies because high concentrations of solid sub-23 nm particles have been found. For example, some investigators found that in specific engine operation modes the majority of the particles can be close to the lower legislated size for GDI (Rönkkö et al., 2014), compressed natural gas (CNG) (Alanen et al., 2015) or even diesel vehicles (Lähde et al., 2010). The addition of additives results in a separate solid nucleation mode below 23 nm (Gidney et al., 2010). More details can be found in relevant reviews (Giechaskiel et al., 2014a, 2017). An overview of the last 3–4 years of testing in our laboratory showed that CNG heavy-duty vehicles and mopeds had almost 100% higher > 10 nm emissions than > 23 nm emissions, while the percentages for motorcycles and port fuel injection (PFI) gasoline vehicles were > 60% for typical test cycles (Giechaskiel et al., 2018a). Monitoring and investigations on the topic are on-going.

In 2017, Real Driving Emissions (RDE) testing on the road with Portable Emissions Measurement Systems (PEMS) was introduced for light-duty vehicles type-approval (EU, 2017), with in-service conformity testing to follow in 2020. On-road SPN measurements are conducted directly at the tailpipe. Thus, measured concentrations might not be identical to those measured at the full dilution tunnel by the regulated procedure. Particle number concentration may decrease in the exhaust transfer line connecting the tailpipe to the dilution tunnel through agglomeration as well as particle tube losses through thermophoresis and diffusion (Isella et al., 2008; Giechaskiel et al., 2012a). Furthermore, the determination of the exhaust flow rate, required for the calculation of the emissions at the tailpipe, is associated with a relatively high uncertainty of 3–10% (Giechaskiel et al., 2018a). Pressure pulsation and exhaust gas temperature fluctuations may result in an inaccurate primary dilution factor in tailpipe measurement systems. The different response times of the particle number and the exhaust flow signals, along with small errors in misalignment, further increase tailpipe measurement uncertainty.

Comparisons of measured SPN emissions at the tailpipe and the dilution tunnel are still scarce, e.g. (Giechaskiel et al., 2010a; Khan et al., 2018). A limited number of data were generated during PEMS validations (Giechaskiel et al., 2015). Recently, the PEMS (instrument) measurement uncertainty was evaluated for gaseous pollutants (NOx) (Giechaskiel et al., 2018a). For particle measurements the analysis has only been partially completed. In contrast to the concentration of the regulated gaseous pollutants, particle concentrations can be significantly altered as particles are transported along the transfer line (Isella et al., 2008). This effect needs to be better evaluated. Thus, in addition to the instrument measurement uncertainty, the uncertainty that different sampling locations introduce (i.e., tailpipe versus dilution tunnel) has to be quantified. Additional sources of uncertainty are the test conditions (temperature, pressure, vibrations, etc.). This topic will not be examined here, but it will be addressed at the relevant CEN (Comité Européen de Normalisation) standard (CEN/TC 301, 2018).

The objective of this paper is to compare tailpipe and regulated dilution tunnel measurements with the same laboratory-grade equipment to quantify the sampling “location” (and tailpipe methodology) uncertainty. The focus here is on the regulated SPN size range (particle diameter > 23 nm), but SPN including also smaller particles (> 10 nm) is also examined as such emissions might be regulated in the future. Tests were conducted with diesel, gasoline, and compressed natural gas (CNG) vehicles over cold start cycles to cover a wide range of emission conditions.

2. Methods

2.1. Experimental setup

Fig. 1 shows the experimental setup. The basic idea was to install identical instruments (i.e., same SPN instrument models) both at the tailpipe and the full dilution tunnel for all tested vehicles. The tests were conducted at the Vehicle Emissions Laboratory (VELA 2) of the Joint Research Centre (JRC) (Ispra, Italy). The transfer line (tube) connecting the vehicle (its tailpipe) to the dilution tunnel was approximately 6 m heated to 70 °C; the maximum length allowed by the regulation. Different transfer tubes were used for spark ignition (gasoline, CNG vehicles) and compression ignition (diesel) vehicles. All tests for each vehicle were conducted continuously without measuring other vehicles in between (i.e., the same vehicle was used for 1–2 weeks of testing) to minimize the contamination (history) effect of the sampling lines.

2.2. Vehicles

The tested vehicles fulfilled the Euro 6 standard with one exception (Euro 5b). They covered a wide range of engine capacities and vehicle technologies (compression ignition, spark ignition) with different fuel types (diesel, gasoline, CNG). Details can be found in Table 1.

2.3. Cycle

The test cycle was the World Harmonised Light-duty vehicles Test Cycle (WLTC) with engine cold start. The tests were conducted at ambient temperature of 23 °C after soaking (leaving them to stabilize) at 23 °C for at least 12 h. Two to four test repetitions were conducted for each vehicle.
Due to the lack of oil and/or coolant temperatures for all vehicles, we considered as the cold start part of the WLTC the first 300 s of the cycle, in agreement with the RDE regulation (EU, 2017). At 300 s the oil temperature was around 45 °C for the compression ignition vehicles and 55–60 °C for the spark ignition vehicles, thus lower than what is typically considered warm engine (oil or coolant temperature > 70 °C).

2.4. Cold start

The dilution, which includes particle losses as an average of 30 nm, 50 nm and 100 nm, the so called Particle number Concentration Reduction Factor (PCRF\(_{\text{ave}}\)), was approximately 1000 (100 × 10). Downstream the diluter a butanol Condensation Particle Counter (CPC) (model TSI 3779) with 50% counting efficiency at 23 nm (d\(_{50\%}=23\) nm) measured solid particles (Giechaskiel et al., 2009; Wang et al., 2010). In parallel, a butanol CPC (model TSI 3772) (d\(_{50\%}=10\) nm) was used to investigate the presence of particles of diameter between 10 and 23 nm (Takegawa and Sakurai, 2011). An identical combination of instruments was connected to the tailpipe with a 0.5 m heated line at 120 °C, and a final dilution in a porous diluter with room-temperature filtered air. The system had been calibrated by the manufacturer. The dilution, which includes particle losses as an average of 30 nm, 50 nm and 100 nm, the so called Particle number Concentration Reduction Factor (PCRF\(_{\text{ave}}\)), was approximately 1000 (100 × 10). Downstream the diluter a butanol Condensation Particle Counter (CPC) (model TSI 3779) with 50% counting efficiency at 23 nm (d\(_{50\%}=23\) nm) measured solid particles (Giechaskiel et al., 2009; Wang et al., 2010). In parallel, a butanol CPC (model TSI 3772) (d\(_{50\%}=10\) nm) was used to investigate the presence of particles of diameter between 10 and 23 nm (Takegawa and Sakurai, 2011). An identical combination of instruments was connected to the tailpipe with a 0.5 m heated line at 120 °C. The PCRF\(_{\text{ave}}\) in the tailpipe was 2000 (200 × 10). We used identical instruments to minimize the parameters that can have an effect on the emission measurements, focusing on differences originating from the location of the instruments. Previous studies suggest that both solid and volatile artefacts appear in SPN systems when 10 nm CPCs are connected; for this reason catalytic strippers are recommended for sub-23 nm measurements (Giechaskiel et al., 2014a). In our case we used high primary dilution to minimize such artefacts (Yamada et al., 2015).

The two sets of instruments were compared to each other at the dilution tunnel measuring simultaneously vehicle CNG #2. The "tailpipe" instruments measured 5–7% less; this difference was not taken into account in our results.

An Engine Exhaust Particle Sizer (EEPS 3090) (model 3090 from TSI, Shoreview, MN, USA) (Johnson et al., 2004) was connected to the full dilution tunnel via an AVL catalytic stripper (Amanatidis et al., 2013) to measure the size distribution (5.6–560 nm) of solid particles. Filtered air was added downstream the catalytic stripper to reach the flow rate required by the EEPS. The PCRF\(_{\text{ave}}\) was around 10. In this case the catalytic stripper was necessary because any other thermal pre-treatment system would require high dilution and lower the concentrations below the detection limit of the instrument. EEPS measurements are based on charging the particles, applying an electric field and measuring their current at the electrometers where they deposit. The fractal (soot) inversion algorithm was applied (Wang et al., 2016).

The exhaust flow rate was estimated as the difference of the total flow of the full dilution tunnel (CVS) minus the airflow entering into it. Calibration and adjustment of the intake air to the CVS flow was conducted before each test. For a few vehicles (PFI #1, DPF #2), an AVL exhaust flow meter was connected to the tailpipe of the vehicle to compare the two exhaust-flow measurement methods.

2.6. Calculations: measurement corrections, uncertainties, and particle losses

2.6.1. Sub-23 nm emissions

The particle number concentrations > 23 nm (SPN\(_{23}\)) reported by the instruments were already corrected for the PCRF\(_{\text{ave}}\), the slope factor of the CPC, and normalized to 0 °C and 101.3 kPa. To take into account the higher losses in the sub-23 nm range, the > 10 nm concentrations (SPN\(_{10}\)) were corrected to obtain an improved estimate of the true > 10 nm concentrations (SPN\(_{10,\text{corr}}\)) as follows (Giechaskiel et al., 2018a):

\[
\text{SPN}_{10,\text{corr}} = \text{SPN}_{10} / \text{PCRF}_{10/100}\times \text{PCRF}_{\text{10/100}}
\]

This correction assumes that the particles in the 10–23 nm range can be corrected by the particle losses of the 15 nm size, which was available from the instruments calibrations.

The EEPS data were corrected for size dependent particle losses in the catalytic stripper by the catalytic stripper penetration curve given in Amanatidis et al. (2013).

2.6.2. Dilution tunnel emissions

The SPN\(_{\text{CVS}}\) emissions [p/km] were calculated by multiplying the volume of the diluted exhaust gas V\(_{\text{CVS}}\) [l] (normalized to 0 °C and 101.3 kPa) by the concentration C\(_{\text{CVS}}\) [p/cm\(^3\)] during the appropriate time interval (normalized to 0 °C and 101.3 kPa and corrected for the PCRF\(_{\text{ave}}\)) and the slope of the CPC (EU, 2017), followed by a division by the kilometres covered (D):

\[
\text{SPN}_{\text{CVS}} = V_{\text{CVS}} C_{\text{CVS}} 1000 / D
\]

The uncertainty can be estimated from the error propagation rule:

\[
\sigma^2(\text{SPN}_{\text{CVS}}) = \sigma^2(V_{\text{CVS}}) + \sigma^2(C_{\text{CVS}}) + \sigma^2(D)
\]

The uncertainty of the final result (9–18%) is dominated by the uncertainty of the particle counter uncertainty (9–18%) because the volume uncertainty (2%) and the distance uncertainty (< 0.5%) are relatively small. The volume uncertainty and the distance uncertainties were taken as the maximum error permitted in the regulation at the annual calibration of the instrument (EU, 2017). The particle counter uncertainty was based on the uncertainty of the instruments used for calibrating the particle number systems (Giechaskiel et al., 2012b). The same uncertainty was assumed for both > 23 nm and > 10 nm measurements, although for size distributions peaking below 40 nm the > 10 nm systems uncertainty may be higher due to the higher uncertainties in the penetration curves of the commercial systems (Giechaskiel and Martini, 2014; Giechaskiel et al., 2017).

2.6.3. Tailpipe emissions

The instantaneous particle number flow rate SPNI [p/s] was determined by multiplying the instantaneous particle number concentration C\(_{\text{SPN,I}}\) [p/cm\(^3\)] (normalized to 0 °C and 101.3 kPa) by the
instantaneous exhaust mass flow rate $q_{exh}$ [kg/s] (aligned to each other), and by dividing the product by the density of the exhaust gas $\rho_{exh}$ [kg/m$^3$] at 0 °C (EU, 2017). A density of 1.2943 was used for diesel (B7), 1.2661 for CNG, and 1.2931 kg/m$^3$ for gasoline (E10). The total SPN emissions were calculated by integrating the SPN rate over the appropriate time interval (e.g., the first 300 s-cold start or the complete cycle of 1800 s) and by dividing it by the appropriate distance covered $D$ (around 2 km or 23.2 km respectively). Specifically,

$$SPN_{TP} = \sum SPN_i / D$$

As before, the uncertainty can be estimated from the error propagation rule. In addition we assumed, as a first approach, that the uncertainty at different concentrations or exhaust flow rates remains the same (i.e., it is independent of the measured concentrations):

$$\sigma^2(\text{SPN}_{TP}) = \sigma^2(q_{exh}) + \sigma^2(C_{SPN}) + \sigma^2(D)$$

The uncertainty of the final result (10–21%) depends mainly on the uncertainty of the particle counter (9–18%) and the uncertainty of the exhaust flow rate (3–10%) (Giechaskiel et al., 2018a). The dynamics of the two signals and the time misalignment (5%) also contribute to the final error: they may increase the final uncertainty to 15–26%. To the best of our knowledge studies that analyse SPN measurement uncertainties do not exit: the numbers reported here are only indicative.

2.6.4. Theoretical analysis: particle losses

We estimated particle losses from the tailpipe to the full dilution tunnel by considering agglomeration, thermophoresis, and diffusion of the most relevant aerosol processes (Isella et al., 2008; Giechaskiel et al., 2012a). A detailed, experimental and theoretical analysis of particle transport and losses in the transfer tube was performed elsewhere (Isella et al., 2008). Their estimates of the relative importance of these three processes, in addition to the particle residence time in the transfer tube, support this choice.

Thermophoretic deposition was calculated by taking as input temperature the exhaust gas temperature at the tailpipe, while the temperature of the heater setting of the transfer tube was taken to be the exit temperature (70 °C). We assumed that no thermophoretic deposition occurred during dilution in the dilution tunnel as the temperature drop of the mixture is small there.

The effect of agglomeration on the particle size distribution was calculated every second by taking the measured concentration at the tailpipe as the inlet concentration with a mean residence time. The mean particle residence time in the transfer tube was calculated from the volume of the tube and the mean exhaust flow rate during the first 300 s. The average agglomeration coefficient of a polydisperse size distribution with Geometric Mean Diameter (GMD) 30 nm was taken to be $1.1 \times 10^{-15}$ m$^3$/s (Giechaskiel et al., 2012a).

In addition to particle losses, the effect of the extracted flow from the tailpipe was analysed. The extraction of a flow from the tailpipe results in higher dilution in the dilution tunnel. This higher dilution is usually not taken into account in calculations of particle number concentrations by the laboratory automation systems. In our case, as we compare tailpipe to dilution tunnel measurements, the extracted flow dilution has to be considered. The concentrations at the dilution tunnel were recalculated every second by using the “true” dilution as if no flow had been extracted.

Typical values used in the calculation of the effect of various aerosol processes on the particle concentration in the transfer tube (under conditions of the cold start part of the WLTC cycle) are summarized in Table 2.

Finally, the effect of misalignment of the exhaust flow rate and particle number concentrations was estimated by recalculating tailpipe emissions via shifting the SPN signal by $\pm 1$ s.

The fraction of sub-23 nm particles, measured with the CPCs, was also plotted as a function of the mean GMD estimated by the EEPS for the cold start part of the WLTC (corrected for the losses in the catalytic stripper). In this comparison all measurements with concentrations $< 10^7$ p/km$^3$ were excluded to eliminate the uncertainty arising from the lower detection limit of the EEPS. The expected sub-23 nm fraction as a function of the GMD was calculated using the 23 nm and 10 nm CPC concentrations, the penetration efficiencies of the APC, and the detection efficiencies of the CPCs, as reported in the literature (Giechaskiel et al., 2009, 2017; Takegawa and Sakurai, 2011).

3. Results

3.1. Emission levels: CVS measurements

The emissions levels of the various vehicles over the WLTC are summarized in Fig. 2 (upper panel). The error bars (one standard deviation of two to four repetitions) are given only for the 23 nm measurements for better readability of the figure. Error bars for the 10 nm measurements were similar. The SPN limit $6 \times 10^{11}$ p/km for sizes $> 23$ nm is applicable to diesel vehicles after September 2011 and to GDIIs after September 2017. Diesel vehicle emissions are well below the limit; the DPF-equipped diesels are the lowest emitting vehicles in the group of vehicles examined. The emissions of the GDI vehicle, even though not equipped with a Gasoline Particulate Filter (GPF), are 3 times below the limit. The emissions of one of the CNG vehicles are close to the limit, while those of the other well below it. The emissions of gasoline vehicles with Port Fuel Injection (PFI) are higher than those of GDI vehicles, with one of them exceeding the limit by a factor of five. Note, however, that there is no regulated limit for PFI or CNG vehicles, yet.

The percentage of measured particles between 10 nm and 23 nm (with respect to the $> 23$ nm particles) is 26–31% for the diesel vehicles, 127–152% for the CNG vehicles, 74% for the GDI, and 14–47% for the PFIs. Only in one case inclusion of sub-23 nm particles would have resulted in exceedance of the limit (CNG #1). Note the logarithmic y-axis.

During cold start (first 300 s) emissions are higher than for the whole cycle (Fig. 2 lower panel), but the sub-23 nm fractions remain at similar levels, with the exception of two diesel vehicles for which the fraction increased.

3.2. Tailpipe versus dilution tunnel

Fig. 3 shows the percentage differences of tailpipe (TP) from dilution tunnel (CVS) measurements (with respect to dilution tunnel measurements) for the complete cycle (upper panel) and for the cold-start part (lower panel). For the complete cycle (Fig. 3, upper panel) the differences are in general within 15% with one exception (PFI #2) that reached 25% in the 10 nm tests. For the cold-start part of the cycle (Fig. 3, lower panel), differences are higher exceeding 30% for the PFIs.

4. Discussion

4.1. Emission levels (CVS)

The emission levels measured in this study are generally in agreement with levels reported in the literature. The diesel vehicles equipped with Diesel Particulate Filters (DPF) had emission levels lower than the current limit, as typically reported, for example, for Euro 5 (Mamakos et al., 2013a; Bielaczyc et al., 2014) and Euro 6 (Mamakos et al., 2013b, Bielaczyc et al., 2017; Suarez-Bertoa and Astorga, 2018; Giechaskiel et al., 2018a) vehicles. PFI vehicle emissions depended on the vehicle tested, being lower than the current limit for one vehicle (PFI#1), and higher for the other (PFI#2). Such PFI vehicle-dependent SPN emissions lower or higher than the (diesel) SPN limit have been measured before for China 4 (He et al., 2018), Euro 5 (Bielaczyc et al., 2015), or even Euro 6b (Hensel et al., 2018) vehicles. The SPN emissions of the
CNG vehicles varied but they were below the (diesel) SPN limit. The CNG vehicle with emissions close to the limit had only 1000 km on the odometer; thus, it is possible that the emissions were high due to the high contribution of the lubricant. Others have also reported emissions from CNG vehicles below the limit for Euro 5 (Bielaczyc et al., 2014, 2015), and Euro 6 vehicles (Bielaczyc et al., 2017) or around the limit for a Euro 4 vehicle (Schreiber et al., 2007).

What is rather new is that the GDI vehicle without Gasoline Particulate Filter (GPF) had much lower emissions than the limit: they were even lower than the emissions from the PFIs. A few years ago it was demonstrated that the regulated SPN 23 nm limit could be fulfilled via internal engine measures and injection strategies optimized for specific driving cycles (McAllister et al., 2014). However, we are not aware of studies reporting low emissions for commercially available vehicles without GPF. A recent study showed that the emission levels of a commercially available GDI vehicle without GPF were at or slightly higher than the limit (Demuynck et al., 2017). We did not test the vehicle in our study under different cycles to ascertain whether it has low emissions under all driving conditions. Most recent studies with China 4 (Zhu et al., 2016; He et al., 2018), Euro 5 (Bielaczyc et al., 2014; Mamakos et al., 2013b), even Euro 6 (Bielaczyc et al., 2017; Suarez-Bertoa and Astrorga) GDI vehicles measured emissions higher than the limit. According to literature studies, emissions decreased only when GPF had been installed (Mamakos et al., 2013a; Czerwinski et al., 2017; Giechaskiel et al., 2018a; Joshi and Johnson, 2018).

4.2. Cold start (CVS)

The emissions during the cold start section of the cycle were high. This arises from incomplete combustion and higher lube oil contribution when the engine is cold, and the short distance travelled in the first 5 min (around 2 km). Only one CNG vehicle (CNG #2) and the two Euro 6 DPF vehicles emitted less than $6 \times 10^{11}$ p/km (> 23 nm) for the WLTC. Sub-23 nm SPN in red, > 23 nm in blue. Logarithmic y-axis. Error bars (23 nm measurements) show one standard deviation from 2 to 4 repetitions.

### Table 2

| Code     | PFI #1 | PFI #2 | GDI | CNG #1 | CNG #2 | DPF #1 | DPF #2 | DPF #3 |
|----------|--------|--------|-----|--------|--------|--------|--------|--------|
| Extracted Flow (l/min) | 24     | 9      | 20  | 40     | 15     | 21     | 9      | 24     |
| Residence Time (s)     | 7.3    | 7.5    | 9.5 | 3.5    | 8.2    | 3.4    | 3.8    | 4.6    |
| Peak tailpipe SPN$_{23}$ ($\times 10^6$ p/cm$^3$) | 54     | 68     | 10  | 5      | 0.13   | 0.24   | 0.40   | 5.6    |
| $T_{\text{cold}}$ (°C) | 71     | 70     | 50  | 220    | 65     | 40     | 52     | 47     |

### Fig. 2

Solid Particle Emissions (SPN) during the WLTC (upper panel) and the first 300 s of the WLTC (lower panel) for the various vehicles. A SPN limit of $6 \times 10^{11}$ p/km (> 23 nm) for the WLTC is applicable to diesel (after September 2011) and GDI vehicles (after September 2017). Sub-23 nm SPN in red, > 23 nm in blue. Logarithmic y-axis. Error bars (23 nm measurements) show one standard deviation from 2 to 4 repetitions.

### Fig. 3

Percentage difference of tailpipe (TP) from dilution tunnel (CVS) measurements (with respect to CVS measurements) for the complete cycle (upper panel) and the cold-start part (lower panel). Error bars show one standard deviation of 2 to 4 repetitions.

Bertoa and Astrorga GDI vehicles measured emissions higher than the limit. According to literature studies, emissions decreased only when GPF had been installed (Mamakos et al., 2013a; Czerwinski et al., 2017; Giechaskiel et al., 2018a; Joshi and Johnson, 2018).

4.3. Sub-23 nm particles: tailpipe versus dilution tunnel measurements

The percentage of sub-23 nm particles (relative to the > 23 nm measurement) not regulated today was around 30% for the diesel vehicles of this study, up to 74% for the gasoline vehicles, and > 125% for the CNG vehicles. These finding are in agreement with our previous findings and literature studies (Mamakos et al., 2013a, 2013b;
EEPS was connected to the dilution tunnel. Refers to measurements at the dilution tunnel, while TP at the tailpipe. The calculated excess concentrations for different Geometric Mean Diameters (GMDs) measured by the EEPS during the cold start part of the WLTC. CPCs (see Section Methods). The sub-23 nm percentages at the dilution tunnel systems as a function of the GMD estimated by the EEPS are considered. High soot emissions of the specified vehicle, which resulted in a low relative percentage of sub-23 nm particles, typically originating from the lubricant. The high sub-23 nm percentages at the cold start part of two of the DPF vehicles (actually only during the first minute) are quite puzzling. For DPF #1 the concentrations were too low to draw definitive conclusion. For DPF #2 the EEPS with thermal pre-treatment of the sample in a catalytic stripper also showed high number of sub-23 nm particles. A test with hot engine start (not reported herein) did not show this high percentage. Thus, the high sub-23 nm emissions are related to the formation of particles during the first minute: probably these particles are less graphitized and shrink in the thermal pre-treatment systems. Fig. 4 plots the solid-particle size distributions during the cold start of the WLTC for the various vehicles. Only concentrations above the detection limit of the EEPS are considered.

Fig. 5 shows the percentage of sub-23 nm particles based on the difference between the 10 nm and 23 nm CPCs for both tailpipe and dilution tunnel systems as a function of the GMD estimated by the EEPS (connected to the dilution tunnel). The dotted lines show the percentages calculated via the penetration curves of the SPN system and the CPCs (see Section Methods). The sub-23 nm percentages at the dilution tunnel and the tailpipe are quite close to each other, with a tendency of higher values at the tailpipe at smaller sizes. It is not clear whether this trend of higher tailpipe sub-23 nm concentrations (in absolute terms and with respect to CVS concentrations) with decreasing GMD diameter is due to experimental uncertainty or a real effect. We think it is a real effect: particles in the dilution tunnel are slightly bigger due to agglomeration in the transfer tube. For smaller particles the agglomeration coefficient is higher, and thus their growth more significant. At the 10–25 nm size range the penetration efficiency of the instruments is also very sensitive to particle size as it decreases steeply, thus any size change is more evident. Another possibility is that the removal of material adsorbed on the particles from the dilution tunnel system is not as efficient as it is when sampling occurs directly from the tailpipe under hot conditions. We note relatively good agreement between theory and experiments for the dilution tunnel results, indicating that the information the two CPC counters provide could be used to obtain size information, in addition to the very important information on the actual sub-23 nm emission levels. Although the chemical composition of such particles is not well known, recent studies suggest that they may consist predominantly of ash (Liati et al., 2018). There are concerns that sub-23 nm particles may be even more harmful than bigger particles due to their larger surface area for the same mass (surface area to volume ratio) and, thence, their higher bioactivity/surface reactivity (Dick et al., 2003; Sager and Castranova, 2009).

4.4. Differences between tailpipe and dilution tunnel measurements

The results of tailpipe and dilution tunnel measurements, Fig. 3, are in good agreement for the complete cycles, within 15% (tailpipe higher). Tailpipe results were lower only in one case (DPF #1) where the emission levels were close to the detection limit of the dilution tunnel instruments. The differences at the cold-start part were higher, reaching 35% for the PFI cases. The dilution tunnel measurements are often considered to depend only on the uncertainty of the SPN system. As mentioned earlier, the SPN systems at the dilution tunnel and the tailpipe were once compared while sampling together from the dilution tunnel (vehicle CNG #2). The difference was < 7% (tailpipe-measuring system lower). Thus, other uncertainties were investigated to understand the differences between tailpipe and dilution tunnel measurements.

Tailpipe measurements are subject to exhaust flow uncertainty; based on comparisons with PEMS exhaust flow meters this uncertainty was estimated to be around ± 6%, in line with reported differences in the literature, 3–10% (Giechaskiel et al., 2018a; Varella et al., 2018). The time misalignment of the exhaust flow and SPN signals of ± 1 s was determined to introduce an uncertainty of around ± 5%. However, in both cases, no particular positive or negative trend was found.

The differences were further assessed by performing a theoretical analysis of particle losses between the tailpipe and the full dilution tunnel during cold start (Table 3) (see Section Methods). Thermophoretic losses during cold start should be negligible because the exhaust gas temperature at the tailpipe sampling location is < 50–70 °C for the first 300 s (except for CNG #1 < 220 °C). Thermophoresis can play an important role only at the high-speed part of the cycle (< 4% in the examined cases). Diffusional losses were estimated to be 1% (23 nm) to 3% (10 nm). Agglomeration was found to be significant reaching 17% for the vehicles with high cold start emissions (Table 3).

Another source of error of the SPN dilution tunnel measurements is that any extracted sample from the tailpipe results in higher dilution at the full dilution tunnel, and, consequently, to lower concentrations. The effect is small (< 3% even for the 1.21 gasoline engine) when < 10 l/min are sampled, but it can become significant when many analysers are connected leading up to 8% error (24 l/min, 1.4 l gasoline engine). Today, most laboratory automation systems do not consider this error for particle number systems: this may improve in the future.

Therefore, the calculations show that corrections for particle losses bring the tailpipe results within 10% of the dilution tunnel results, even for the cold-start part, a value that is within the measurement.
uncertainty of the SPN systems. This point needs special attention. Tailpipe measurements are associated with slightly higher uncertainty (theoretically 15–26% vs. 9–18% at the dilution tunnel, see Section Methods). Yet, our analysis shows that the main reason of the differences between tailpipe and dilution tunnel measurements is the processes that occur between the tailpipe and the full dilution tunnel, i.e., along the transfer line (for a theoretical analysis, see Isella et al., 2008). Thus, these differences are due to the current regulated experimental methodology. Improving it would require heating the tubing and shortening the length of the transfer line. However, there are practical restrictions on the chassis dynamometers that would render very difficult to reduce the 6 m length limit of the transfer line connecting the vehicle tailpipe to the dilution tunnel. Corrugated tubes could further increase losses via deposition. Dilution tunnels could also introduce additional losses during the mixing of the exhaust gas with the dilution air; however, these losses haven’t been characterized. In general, inter-laboratory correlation exercises give a reproducibility of 9–29% for modern vehicles, which is higher than the theoretically expected (9–18%) (Giechaskiel et al., 2018c). The important point is that “true” cold start emissions (i.e., what would be emitted in the atmosphere) are those measured at the tailpipe. The dilution tunnel (CVS) methodology slightly underestimates them. This tailpipe-CVS measurement difference should also be taken into account when PEMS emissions are estimated. This tailpipe-CVS measurement difference was characterized. In general, inter-laboratory correlation exercises give a reproducibility of 9–29% for modern vehicles, which is higher than the theoretically expected (9–18%) (Giechaskiel et al., 2018c). The important point is that “true” cold start emissions (i.e., what would be emitted in the atmosphere) are those measured at the tailpipe. The dilution tunnel (CVS) methodology slightly underestimates them. This tailpipe-CVS measurement difference should also be taken into account when PEMS emissions are compared to SPN emissions at the dilution tunnel. It should be mentioned that agglomeration losses with the current emission levels (< 1 × 10^12 p/km) are much lower compared to losses for older vehicles with 1–2 orders of magnitude higher emissions (17% in this study vs. 40% in Isella et al., 2008).

Finally, direct tailpipe measurements have additional advantages: they need less space, as there is no need for big full dilution tunnel installations nor for proportional partial flow systems, as currently required by the light- and heavy-duty emission regulation. Most importantly, type-approval measurements in the laboratory would be directly comparable to PEMS measurements on the road. The feasibility of tailpipe measurements for type approval of engines will be investigated by the PMP group in the future for post Euro 6/VI regulations. In particular, the focus at the moment is on heavy-duty vehicles, where the focus is on heavy-duty engines, where temperature variations, particle concentrations, and pressure fluctuations are larger. Thus, the conclusions of this study for light-duty vehicles cannot be extrapolated to heavy-duty engines.

Finally, we stress that we used identical instrumentation at the tailpipe and the dilution tunnel because the focus of our study was on the effect of instrument location (and methodology) on SPN results. We also investigated measurement repeatability at the tailpipe and the dilution tunnel because the focus of our study was on the effect of instrument location (and methodology) on SPN results. We also investigated measurement repeatability at the tailpipe and the dilution tunnel with the 23 nm and 10 nm systems. We found that particle concentrations at the dilution tunnel were lower due to changes that occurred in the transfer line, the tube that connects the tailpipe to the dilution tunnel, and the dilution tunnel itself (up to 17% at cold start in our study). We identified agglomeration as the dominant mechanism (more than thermophoretic or diffusional losses) responsible for the particle concentration decrease.

We evaluated the particle number measurement uncertainty in emission measurements of eight vehicles of different technologies and engine capacities under the World Harmonised Light-duty Test Cycle. The objective was to estimate the effect of instrument location (tailpipe versus dilution tunnel) and methodology on Solid Particle Number (SPN) measurements. Identical instruments were used at both the tailpipe and the dilution tunnel measurement points. Concentrations of particles with diameter greater than 23 nm, as required by current regulations, and greater than 10 nm, as envisaged for future regulations, were measured.

The overall result is that tailpipe measurements do not significantly increase the measurement uncertainty with respect to dilution tunnel measurements. The uncertainty of the exhaust flow rate determination and the time misalignment can contribute around 10% and 5% respectively. We found that particle concentrations at the dilution tunnel were lower due to changes that occurred in the transfer line, the tube that connects the tailpipe to the dilution tunnel, and the dilution tunnel itself (up to 17% at cold start in our study). We identified agglomeration as the dominant mechanism (more than thermophoretic or diffusional losses) responsible for the particle concentration decrease.

Particle concentrations below 23 nm were high in some cases, exceeding the limits imposed by the current regulation. These emission results indicate that decreasing the 23 nm lower particle size should be seriously considered in future regulations. We found that the repeatability of the > 10 nm measurements was the same or better to the > 23 nm measurements both at the dilution tunnel and at the tailpipe. We note, however, that systems with different size-dependent losses could

Table 3

| Code         | PFI #1 | PFI #2 | GDI | CNG #1 | CNG #2 | DPF #1 | DPF #2 | DPF #3 |
|--------------|--------|--------|-----|--------|--------|--------|--------|--------|
| Measured difference | 34.0%  | 28.2%  | 5.1% | 3.7%   | 14.3%  | 0.6%   | 3.4%   | 4.2%   |
| Calculated difference | 30.4%  | 18.3%  | 8.4% | 5.1%   | 6.9%   | 1.6%   | 1.0%   | 4.2%   |
| Agglomeration losses | 16.6%  | 15.6%  | 2.7% | 0.4%   | 0.6%   | 0.0%   | 1.0%   | 1.2%   |
| EFM difference | 5.8%   | 5.8%   | 5.8% | 4.7%   | 6.3%   | 1.6%   | 3.0%   | 3.0%   |
| Extracted flow | 8.0%   | 2.7%   | 5.7% | 5.7%   | 5.7%   | 5.7%   | 5.7%   | 5.7%   |

Estimated uncertainties and particle losses from the tailpipe to the dilution tunnel based on actual tailpipe data. Cold start part of the WLTC cycle, Fig. 3 (lower panel), > 23 nm measurements. EFM stands for Exhaust Flow Measurements.
result in higher differences at the > 10 nm measurements: the effect, though, will be similar for dilution tunnel and tailpipe measurements.

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Conflicts of interest
The authors declare no conflict of interest.

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References
Aiken, J., Sautko, E., Lehtoranta, K., Murtonen, T., Timonen, H., Hillamo, R., Karjalainen, P., Kukulainen, H., Hara, J., Keskinen, J., Rininkö, T., 2015. The forma-

tion and physical properties of the particle emissions from a natural gas engine. Fuel 162, 155–161.

Amanatidis, S., Nitzhachristos, L., Giechaskiel, B., Katsosannis, D., Samaras, Z., Bergmann, A., 2013. Evaluation of an oxidation catalyst (“catalytic stripper”) in eliminating volatile material from combustion aerosol. J. Aerosol. Sci. 57, 144–155.

Andersson, J., 2018. Measuring automotive exhaust particles down to 10 nm. In: Proceedings of the 48th meeting, Down To Ten project presentation to the PMP group, 7 November 2018, Ipsra, Italy.

Bielaczyc, P., Szczotka, A., Woodburn, J., 2015. Regulated and unregulated exhaust emissions from CNG fueled vehicles in light of Euro 6 regulations and the new WLTP/ GTR 15 test procedure. SAE Int. J. Engines 6 (3), 1300–1312.

Bielaczyc, P., Szczotka, A., Woodburn, J., 2017. Investigations into exhaust particulate emissions from multiple vehicle types running on two chassis dynamometer driving cycles. SAE Technical Paper 2017-01-1007, 2017, https://doi.org/10.4271/2017-01-1007.

Bielaczyc, P., Woodburn, J., Szczotka, A., 2014. Particulate emissions from European vehicles featuring direct injection spark ignition engines tested under laboratory conditions. SAE Int. J. Fuels Lubr. 7 (2), 580–590. https://doi.org/10.4271/2014-01-1608.

CEN/TC 301, 2008. Road vehicles. Portable emission measuring systems (PEMS) per-
formance assessment. Proposal for European Standard 2018.

Chen, L., Liang, Z., Zhang, X., Shuai, S., 2017. Characterizing particulate matter emissions from GDI and PFI vehicles under transient and cold start conditions. Fuel 199, 131–140.

Czerwinski, J., Comte, P., Heeb, N., Mayer, A., Hensel, V., 2017. Nanoparticle emissions of DI gasoline cars with/without GPF. SAE Technical Paper 2017-01-1004, 2017, http://dx.doi.org/10.4271/2017-01-1004.

Demuyck, J., Favre, C., Bostees, D., Hamje, H., Andersson, J., 2017. Real-world emis-
sions measurements of a gasoline direct injection vehicle without and with a gasoline particulate filter, SAE Technical Paper 2017-01-0985, 2017.

Dick, G., Brown, D., Donahue, K., Stone, V., 2001. The nucleation of free radicals in the toxicity and inflammatory effects of four ultrafine particle types. Inhal. Toxicol. 15, 39–52.

EU, 2017. Commission Regulation (EU) 2017/1151 of 1 June 2017 supplementing reg-
ulation (EC) No 715/2007 of the European Parliament and of the council on type-
approval of motor vehicles with respect to emissions from light passenger and commercial vehicles (Euro 5 and Euro 6) and on access to vehicle repair and main-
tenance information, amending Directive 2007/46/EC of the European Parliament and of the council, Commission Regulation (EC) No 692/2008 and Commission Regulation (EU) No 1230/2012 and repealing Commission Regulation (EC) No 692/ 2008. Off. J. Eur. Union 2017 (L175), 1–732.

Gidney, J., Twigg, M., Kittelson, D., 2010. Effect of organometalic fuel additives on na-
oparticle emissions from a gasoline passenger car. Environ. Sci. Technol. 44, 2562–2569.

Giechaskiel, B., Carriero, M., Martin, G., Bergmann, A., Pongratz, H., Jörgl, H., Jörgl, H., 2010a. Comparison of particle number measurements from the full dilution tunnel, the tailpipe and partial flow systems. SAE Int. J. Fuel Lubr. 2, 512–530.

Giechaskiel, B., Cosenz, M., Jörgl, H., Bergmann, A., 2010b. Calibration and accuracy of a particle number measurement system. Meas. Sci. Technol. 21, 045102.
ultrafine titanium dioxide. Part. Fibre Toxicol. 6, 15.
Schreiber, D., Forss, A., Mohr, M., Dimopoulos, P., 2007. Particle characterisation of modern CNG, gasoline and diesel passenger cars. SAE Tech (paper 2007-2024-0123).
Suarez-Bertoa, R., Astorga, C., 2018. Impact of cold temperature on Euro 6 passenger car emissions. Environ. Pollut. 234, 318–329.
Takegawa, N., Sakurai, H., 2011. Laboratory evaluation of a TSI condensation particle counter (model 3771) under airborne measurement conditions. Aerosol Sci. Technol. 45 (2), 272–283.
Varella, R., Giechaskiel, B., Sousa, L., Duarte, G., 2018. Comparison of portable emissions measurement systems (PEMS) with laboratory grade equipment. Appl. Sci. 8, 1633.
Wang, X., Caldow, R., Sem, G., Hama, N., Sakurai, H., 2010. Evaluation of a condensation particle counter for vehicle emission measurement: experimental procedure and effects of calibration aerosol material. J. Aerosol Sci. 41, 306–318.

Wang, X., Grose, M., Caldow, R., Osmondson, B., Swanson, J., Chow, J., Watson, J., Kittelson, B., Li, Y., Xue, J., Jung, H., Hu, S., 2016. Improvement of engine exhaust particle sizer (EEPS) size distribution measurement - II. Engine exhaust particles. J. Aerosol Sci. 92, 83–94.
WHO, 2013. Review of Evidence on Health Aspects of Air Pollution - REVIHAAP Project. WHO Regional Office for Europe, Copenhagen, Denmark, pp. 2013.
Yamada, H., Funato, K., Sakurai, H., 2015. Application of the PMP methodology to the measurement of sub-23 nm solid particles: calibration procedures, experimental uncertainties, and data correction methods. J. Aerosol Sci. 88, 58–71.
Zhu, R., Hu, J., Bao, X., He, L., Lai, Y., Zu, L., Li, Y., Su, S., 2016. Tailpipe emissions from gasoline direct injection (GDI) and port fuel injection (PFI) vehicles at both low and high ambient temperatures. Environ. Pollut. 216, 223–234.