

Particle and VOC Emissions from Stoichiometric Gasoline Direct Injection Vehicles and Correlation Between Particle Number and Mass Emissions

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Abstract Particle number (PN) emissions from the following types of passenger cars were compared: normal gasoline direct injection (DI), gasoline DI combined with downsized turbocharged technology, gasoline DI with hybrid technology, gasoline multi-point injection (MPI), gasoline MPI hybrid, and diesel with a diesel particulate filter. Particles emitted from all types of vehicles were mainly over 23 nm in size; the number of sub-23 nm particles was negligible when the internal dilution ratio of a volatile particle remover was 3000, suggesting that the anomalous extra sub-23 nm particles reported to be emitted from gasoline DI cars are semi-volatile particles. Simultaneous measurement of organic species in the exhaust found emission peaks corresponding to the engine start process in the case of gasoline DI hybrid cars. A good two-stage linear correlation between particle mass and PN emissions was observed for gasoline DI and MPI cars, with slopes of $2.0 \times 10^{12}/\text{mg}$ and $2.5 \times 10^{11}/\text{mg}$ below and above 3 mg/km, respectively.

Keywords Gasoline direct injection · Particle number · Proton-transfer-reaction plus switchable reagent ion mass spectrometry · Particle mass · Sub-23 nm particles

1 Introduction

Particulate matter (PM) from automobiles is thought to adversely affect human health, because this PM contains PM_{2.5}, a major component that is clearly related to human diseases [1, 2], as well as ultrafine particles [3], which are considerably more toxic than larger particles [4–6]. Because significant amounts of PM are emitted from diesel engines, the World Health Organization has classified diesel exhaust emissions as a group 1A carcinogen [7].

In the past decade, PM from diesel engines has been reduced remarkably because diesel particulate filters (DPFs), which reduce emissions by more than 90% [8, 9], have been widely adopted, though some issues remain, such as emissions during the regeneration period [10]. The introduction of the particle number (PN) measurement method by the Particle Measurement Programme (PMP) Working Group of the UNECE GRPE [11–15] into exhaust emission regulations [16, 17] played an important role in increasing the adoption of DPFs, because PN measurements are more sensitive than traditional filter weighing and allow governments to set quite low regulation limits that only vehicles with a high-efficiency DPF can pass.

Recently, emission researchers' interest has shifted to PM from gasoline direct injection (DI) engines, as the number of stoichiometric gasoline DI engines is increasing because of their higher thermal efficiency and good compatibility with three-way catalysts. In contrast to traditional multi-point injection (MPI) gasoline engines, this type of engine injects fuel directly into an engine cylinder during the induction stroke. Therefore, the mixing period with fresh air is shorter than in the MPI engine, resulting in higher PM emissions [18–23]. Furthermore, gasoline DI engines are often combined with high-boost

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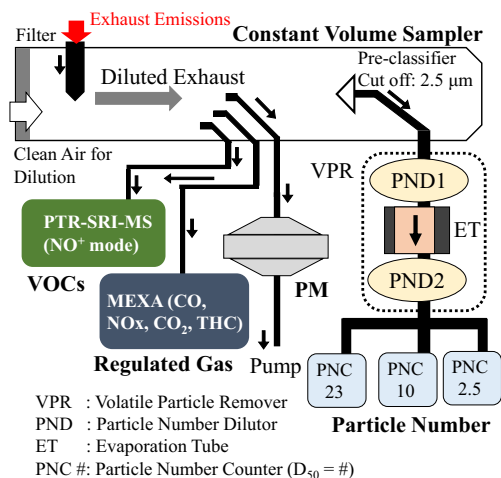


Fig. 1 Outline of measurement systems used in this study

turbocharger or hybrid systems to achieve better fuel economy. Although PM emissions from gasoline DI engines are much lower than those from diesel engines without DPFs, they are much higher than those from diesel engines with DPFs. Thus, the increasing number of stoichiometric gasoline DI engines is having an increasing impact on the roadside environment, and a recent study revealed that PM from gasoline DI engines has a significant effect on the lungs [24]. Against this backdrop, European countries introduced PN regulations for gasoline DI cars from 2014. The limit was 10 times higher than that for diesel passenger cars in the first 3 years but will decrease to a value similar to the one for diesel engines ($6 \times 10^{11}/\text{km}$) in 2017 [25]. A current issue in the UNECE PMP Working Group is whether to lower the D_{50} of the PN measurement method from 23 nm [26]. This is being discussed because some studies have found that

higher fractions of sub-23 nm solid particles are emitted from gasoline DI engines (<60%) than from diesel engines (20%) [27–30]. However, there are still not enough observations of the sub-23 nm fraction of PN from gasoline DI engines to reach a definitive conclusion.

As mentioned above, introducing PN regulations is an effective way to reduce particle emissions from automobiles, but this has been done in only European countries. The USA has decided to introduce stricter PM regulations (1 mg/mile [0.6 mg/km]) for passenger cars from 2025, not by adopting PN measurement but by developing a more sensitive PM weighing method [31]. One big problem with introducing PN regulations is that PM air quality standards all over the world have been set using weight-based units, such as “micrograms per cubic meter.” Thus, introducing PN regulations will result in a discrepancy between emission regulations and ambient air quality standards. To avoid such a situation, it is essential to determine the correlation between particle number and particle mass. There have been some studies of this correlation [32, 33], but a common method has not yet been established.

In this study, we evaluated sub-23 nm solid PN emissions from gasoline DI passenger cars. For the measurements, a particle measurement system that meets European regulatory requirements and particle counters in which D_{50} is lower than 23 nm was used. Seven stoichiometric gasoline DI passenger cars, two MPI cars, and two diesel cars with DPFs were tested. One of the gasoline DI cars and single MPI car were hybrid vehicles. During the measurement, real-time volatile organic compound (VOC) composition analysis was performed using proton-transfer-reaction plus switchable reagent ion mass spectrometry (PTR + SRI-MS). Using the obtained data, we investigated the correlation between PN and PM emission measurements.

Table 1 Specifications of the tested vehicles

Car name	GDI1	GDI2	GDI3	GDI4	GDI1t	GDI2t
Manufacturer	Honda	Nissan	Toyota	Mazda	Subaru	Honda
Engine type	Gasoline DI	Gasoline DI	Gasoline DI	Gasoline DI	Gasoline DI + turbo	Gasoline DI + turbo
Displacement (l)	1.496	1.997	1.998	1.977	1.599	1.496
Aftertreatment device	3 way cat.	3 way cat.	3 way cat.	3 way cat.	3 way cat.	3 way cat.
Vehicle weight (kg)	1190	1660	1510	1460	1530	1650
Regulation	2005	2005	2005	2005	2005	2005
Car name	GDI-h	MPI	MPIh	DPF1	DPF2	
Manufacturer	Toyota	Mitsubishi	Toyota	Mazda	Mazda	
Engine type	Gasoline DI hybrid	MPI	MPI hybrid	Diesel	Diesel	
Displacement (l)	2.493	1.498	1.496	2.188	1.489	
Aftertreatment device	3 way cat.	3 way cat.	3 way cat.	DOC + DPF	DOC + DPF	
Vehicle weight (kg)	1660	1090	1080	1520	1130	
Regulation	2005	2005	2005	2009	2009	

DOC diesel oxidation catalyst, DPF diesel particulate filter

2 Experimental Methods

2.1 Particle Number Measurement Using Chassis Dynamometer

In this study, PN, PM, regulated gases, and VOCs from passenger cars were measured. Details are shown in Fig. 1. The tested car was set on the chassis dynamometer, and all of the exhaust gas was introduced into a constant volume sampler (CVS), where it was diluted with filtered air. Total hydrocarbon (THC), CO, NO_x, and CO₂ were measured using an exhaust measuring system (MEXA-7500-DEGR, Horiba, Ltd., Kyoto, Japan). PM was trapped by a TX40 filter and weighed. These measurement systems meet the requirements of standard approval tests in Japan. Real-time emissions of alkanes, alkenes, aromatics, and dienes with carbon numbers of 4 to 11 were measured using PTR + SRI-MS. Detailed information can be found elsewhere [34–36].

The PNs in different size ranges were measured simultaneously with a PN measurement system (APC, AVL LIST GmbH, Graz, Austria) that meets the requirements of the PMP [16] and with two additional condensation particle counters (CPCs), as in our previous study [37]. Particles were sampled downstream of the CVS. In the APC, volatile particles were removed by a volatile particle remover (VPR), and solid particles with D₅₀ values over 23 nm were counted by a PN counter (PNC). In this study, we denote the PN observed using this PNC by PN₂₃, which indicates the number of solid particles over 23 nm. In addition to the normal PNC, we measured the exhaust of the VPR using two additional CPCs (CPC 3776 and CPC 3772, TSI Inc., MN, USA) whose D₅₀ values were 2.5 and 10 nm, respectively. We denote the PNs obtained by these counters by PN_{2.5} and PN₁₀, indicating the numbers of solid particles over 2.5 and 10 nm, respectively. To minimize the effect of

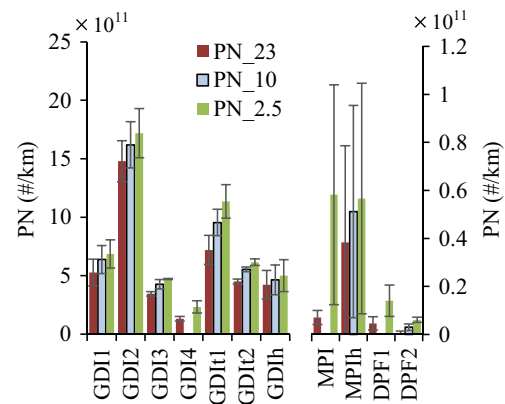


Fig. 3 PN₂₃, PN₁₀, and PN_{2.5} emissions in JC08 hot mode

semi-volatile particle re-nucleation, we set the VPR internal dilution factor at 3000.

2.2 Tested Vehicles and Modes

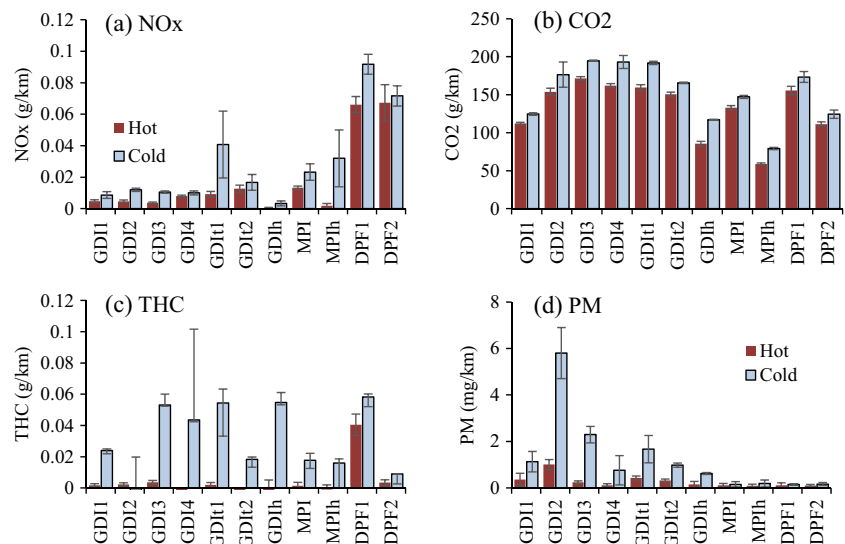
The cars used in this study are listed in Table 1. Seven stoichiometric gasoline DI cars including one hybrid car and two downsized turbocharged cars, two gasoline MPI cars including one hybrid car, and two diesel cars with DPFs were tested. All the cars were passenger cars that passed the most recent exhaust gas regulations in Japan. The test modes used in this study were the JC08 cold and hot modes, which are used in Japanese type approval tests for light-duty cars.

3 Results and Discussion

3.1 Regulated and Particle Number Emissions

The NO_x, CO₂, THC, and PM emissions of all tested cars in the JC08 cold and hot modes are shown in Fig. 2. The tests

Fig. 2 Emissions of **a** NO_x, **b** CO₂, **c** THC, and **d** PM in JC08 cold and hot modes



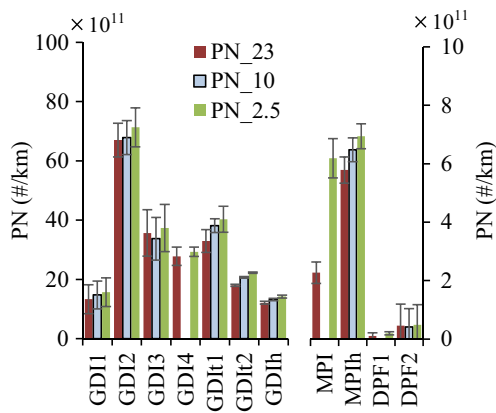


Fig. 4 PN₂₃, PN₁₀, and PN_{2.5} emissions in JC08 cold mode

were carried out at least three times in each condition. In all results, emissions were higher in the cold mode than in the hot mode. NO_x emissions from diesel vehicles were higher than those from the other cars, because neither of the tested diesel cars was equipped with a NO_x aftertreatment device. The hybrid cars, except for MPIh, produced lower NO_x emissions in the cold mode. In the tests, the hybrid cars often ran using only the motor and with the engine stopped, which might have

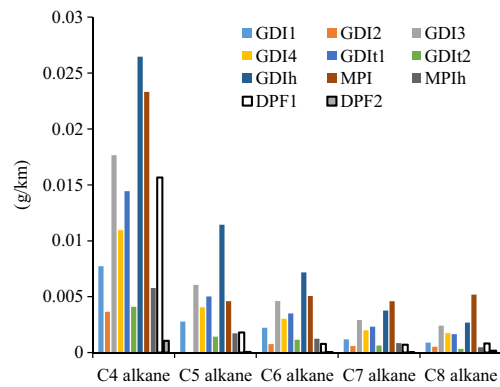
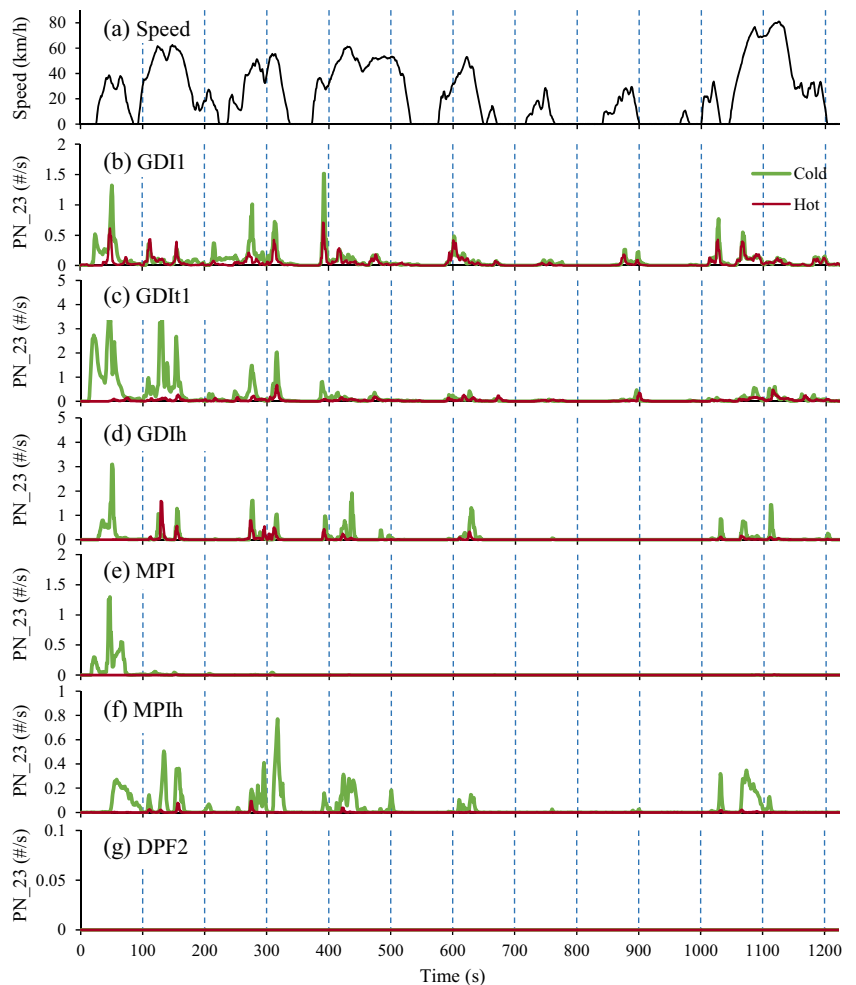


Fig. 6 C4 to C8 alkane emissions in JC08 cold mode. Data were obtained by PTR + SRI-MS

led to lower NO_x emissions. Generally, CO₂ emissions did not depend on the type of engine but did depend on the vehicle weight. However, all the hybrid cars exhibited low CO₂ emissions. PM emissions from all types of gasoline DI cars were higher than those from MPI, MPIh, and DPF diesel cars.

PN₂₃, PN₁₀, and PN_{2.5} in the JC08 hot and cold modes are shown in Figs. 3 and 4, respectively. In the experiments on GDI4, MPI, and DPF1, PN₁₀ was not

Fig. 5 Profiles of **a** speed, and PN₂₃ from **b** GDI1, **c** GDI1t, **d** GDIh, **e** MPI, **f** MPIh, and **g** DPF2 in JC08 cold and hot modes



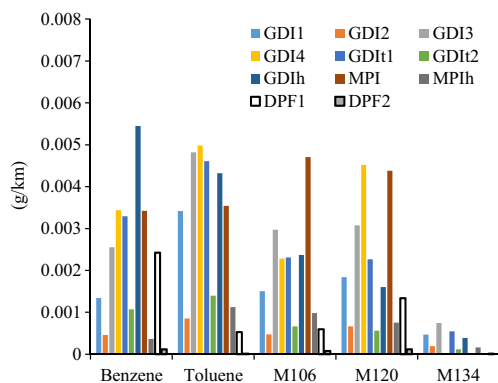


Fig. 7 Aromatics (benzene to M134 species) emissions in JC08 cold mode. Data were obtained by PTR + SRI-MS

measured. PN emissions in the cold mode were almost four times higher than those in the hot mode for gasoline DI cars and 10 times higher for MPI and DPF diesel cars. Similarly, gasoline DI cars produced substantially higher PN emissions than the other cars. In the PM results, a difference between MPI cars and DPF diesel cars was not observed, but in the PN results, a clear difference was observed. PN emissions from gasoline DI cars were similar for normal, downsized turbocharged, and hybrid engines. Common features observed in all cars were that PN_{2.5} was slightly higher than PN₁₀ and that PN₁₀ was slightly higher than PN₂₃. However, these differences were very small. This suggests that PN emissions from all types of engine were mainly due to particles over 23 nm, and the effects of sub-23 nm particles were negligible, even in gasoline DI cars. In this study, the internal dilution factor of the PN measurement system was set to 3000 to avoid the effect of semi-volatile particles. The sub-23 nm particles observed in gasoline DI exhaust by

former studies [27–30] might have been these semi-volatile particles.

Figure 5 shows profiles of PN₂₃ from GDI1, GDI1t, GDIh, MPI, MPIh, and DPF2 in the JC08 cold and hot modes. In all experiments, as discussed above, PN₁₀ and PN_{2.5} profiles were similar to or slightly higher than those of PN₂₃, and anomalous peaks of PN₁₀ and PN_{2.5} were not observed. As shown in Figs. 3 and 4, emissions were higher in the cold mode than in the hot mode in all experiments. Increased emissions were observed in the early part of the test mode, but they disappeared as the engines warmed up. After the engines were completely warmed up, almost no peaks were observed in MPI cars, but still some peaks were observed in gasoline DI cars.

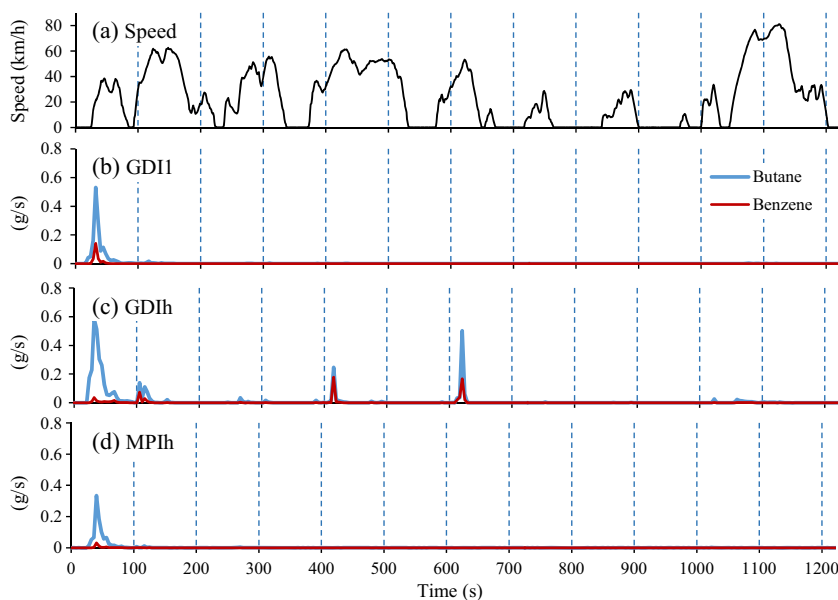
A difference between non-hybrid cars and hybrid cars was observed. Increased emissions corresponding to a cold start in GDI1, GDI1t, and MPI disappeared after about 400 s, and after this, emissions were similar to those of the hot mode. On the other hand, increased emissions in GDIh and MPIh continued until the end of the test. This indicates that hybrid cars need longer to warm up the engine than normal gasoline DI and MPI cars.

PN emissions from the DPF diesels were quite low, and almost no peaks were observed, even in the cold mode.

3.2 VOC Emissions

In this study, we performed real-time VOC species analysis using PTR + SRI-MS. Emissions of alkanes and aromatics in the JC08 cold mode are shown in Figs. 6 and 7, respectively. Differences between the cars were roughly similar to those for THC shown in Fig. 2. C₄ alkane (butane) emissions were the highest, and emissions decreased as the carbon number increased. C₅–C₈ alkane emissions from DPF diesels were relatively low, because diesel fuel contains these alkanes at lower

Fig. 8 a Vehicle speed for JC08 test mode and real-time profiles of butane and benzene from **b** GDI1, **c** GDIh, and **d** MPIh. Data were obtained by PTR + SRI-MS



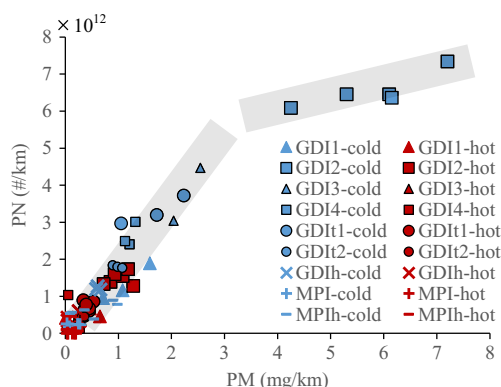


Fig. 9 Correlation between PN and PM in JC08 cold and hot modes from gasoline DI and MPI cars

concentrations than gasoline does. As for aromatics, emissions from M78 (benzene) to M120 (trimethylbenzene, ethylmethylbenzene, or propylbenzene) were similar, even though benzene in fuel is removed because of its effect on health.

Real-time profiles of butane and benzene from GDI1, GDIh, and MPIh in the JC08 cold mode are shown in Fig. 8. All cars, except GDIh, showed similar profiles in which emissions were observed for only the first 100 s. Emissions other than butane and benzene showed similar profiles. On the other hand, GDIh showed a few peaks in the middle of the test. These peaks appeared in timings where the speeds exceeded 50 km/h and may be due to emissions caused when starting to change the power supply from the motor to the engine. However, such peaks were not observed for the hybrid MPI car.

3.3 Correlation Between PM and PN Emissions

Figure 9 shows all PN₂₃ results from gasoline DI and MPI cars in the JC08 cold and hot modes as a function of PM emissions. A clear correlation between PN and PM can be seen. The correlation can be expressed as a two-stage linear shape for which the slope is different below and above 3 mg/km of PM. The slope below 3 mg/km was $2.0 \times 10^{12}/\text{mg}$ and changed to $2.5 \times 10^{11}/\text{mg}$ above 3 mg/km. Using this correlation, the European PN regulation limit for gasoline DI until 2017 ($6 \times 10^{12}/\text{km}$) can be converted to 3 mg/km of PM. This value agrees well with the actual PM limit of 4.5 mg/km. The PN regulation limit of current diesel cars and gasoline DI cars after 2017 is $6 \times 10^{11}/\text{km}$, and the PM value converted from this limit is 0.3 mg/km. This indicates that Europe has already introduced stricter regulations than the USA will introduce in 2025 (1 mg/mile).

4 Conclusions

Particle number and VOC emissions from seven gasoline DI, two gasoline MPI, and two DPF diesel cars were measured.

Two of the gasoline DI cars were downsized turbocharged cars, but no difference from normal gasoline DI cars was observed. One gasoline DI car and one MPI car were hybrid cars, and their CO₂ and NO_x emissions were relatively low.

For PN measurement, solid particles over 23, 10, and 2.5 nm were counted. For all cars, PN₂₃, PN₁₀, and PN_{2.3} were very similar, suggesting that almost all particles were over 23 nm. PN emissions from DPF diesel engines were quite low compared with those from the other types of engines. PN emissions increased at the beginning of the cold start mode and decreased to the level of the hot mode by 400 s in the case of non-hybrid cars. However, increased emissions continued until the end of the test for hybrid cars. The sub-23 nm particles that previous researchers have reported seem to be semi-volatile particles which were removed in this study. VOC emission measurement results obtained by PTR + SIR-MS showed that the main VOC in the exhaust was butane for all cars. Benzene emissions were similar to those for toluene to M120 species, even though its concentration is reduced in fuel for health reasons. VOCs were emitted at the beginning of the test for every car except the gasoline DI hybrid car. The gasoline DI hybrid exhibited some peaks during later stages of the test, possibly due to the power supply changing from the motor to the engine.

The correlation between PM and PN from gasoline DI and MPI cars was good. It was a two-stage linear correlation with slopes of $2.0 \times 10^{12}/\text{mg}$ and $2.5 \times 10^{11}/\text{mg}$ below and above 3 mg/km, respectively. According to this correlation, the PN limit for current diesel and gasoline DI cars after 2017 ($6 \times 10^{11}/\text{km}$) is equivalent to a limit of 0.3 mg/km in terms of PM regulation.

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