


Review

European Regulatory Framework and Particulate Matter Emissions of Gasoline Light-Duty Vehicles: A Review

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Abstract: The particulate matter (PM) emissions of gasoline vehicles were much lower than those of diesel vehicles until the introduction of diesel particulate filters (DPFs) in the early 2000s. At the same time, gasoline direct injection (GDI) engines started to become popular in the market due to their improved efficiency over port fuel injection (PFI) ones. However, the PM mass and number emissions of GDI vehicles were higher than their PFI counterparts and diesel ones equipped with DPFs. Stringent PM mass levels and the introduction of particle number limits for GDI vehicles in the European Union (EU) resulted in significant PM reductions. The EU requirement to fulfill the proposed limits on the road resulted to the introduction of gasoline particulate filters (GPFs) in EU GDI models. This review summarizes the evolution of PM mass emissions from gasoline vehicles placed in the market from early 1990s until 2019 in different parts of the world. The analysis then extends to total and nonvolatile particle number emissions. Care is given to reveal the impact of ambient temperature on emission levels. The discussion tries to provide scientific input to the following policy-relevant questions. Whether particle number limits should be extended to gasoline PFI vehicles, whether the lower limit of 23 nm for particle number measurements should be decreased to 10 nm, and whether low ambient temperature tests for PM should be included.

Keywords: air pollution; vehicle emissions; port fuel injection (PFI); gasoline direct injection (GDI); particle number (PN); particulate matter (PM); low temperature; sub-23 nm; chemical composition

1. Introduction

Driven by the greenhouse gas emission reduction policies and the goal to keep the global temperature increase below 2 °C, the European Union (EU) is committed to reducing CO₂ emissions by 20% in 2020, and by 80–95% in 2050 compared to 1990 levels [1]. The Transport White Paper [2] describes how the transport system can reduce its emissions by 60% in the same period: the target is a 50% shift away from conventionally fueled cars by 2030, phasing them out from cities by 2050. Thus, a big shift to cleaner cars and more sustainable fuels is required. Similar policies are followed globally [3,4]. One contributor in this direction is the replacement of traditional port fuel injection (PFI) gasoline engines by boosted, more efficient, downsized gasoline direct injection (GDI) engines [5].

GDI engines are not new: 100 years ago the very first test rig run of a GDI engine was successfully carried out [6]. Subsequently, this engine was produced for use in boats in the 1930s and for aircrafts in the 1940s. The first serial application in passenger vehicles started in the 1950s [7]. Research into GDIs increased in the 1990s with the development of engine management systems capable of supporting the

additional fuel-metering complexity [8]. The first modern GDI vehicle was introduced in 1996 [9]. The real boost was given later by the European Commission (EC) Recommendation 1999/125/EC to reduce CO₂ emissions from passenger cars. The EU first introduced mandatory 2015 CO₂ standards for new passenger cars in 2009 with Regulation (EC) No 443/2009.

GDI passenger cars for the first time exceeded sales of diesel cars in EU in 2017 (51% vs. 44%) [10]. In the USA GDI cars first penetrated the market in 2007 and light trucks in 2008 [11]. In 2017, the GDI market share (cars and trucks combined) was 50% in the United States (US) with expectations to grow [12].

There have been many review studies on GDI emissions and their control (Table 1). The current study summarizes the regulatory framework: it starts with regulations and shows how and where we have arrived today. It then discusses future perspectives. In contrast to earlier reviews, our study isolates emissions from commercially available vehicles and does not address emissions from engines since only the former can be assumed representative of real-world emissions.

Table 1. Review studies on gasoline engines and their emissions reductions. Y = Yes (addressed).

Review	Health	Engine	Fuel	Aftertreatment	PM/SPN
Book 2009 [13]	-	Y	Y	(Y)	-
Book 2010 [14]	-	Y	Y	(Y)	-
Book 2019 [15]	Y	Y	Y	Y	Y
Book 2019 [16]	Y	Y	Y	Y	Y
Paper 1999 [17]	-	Y	-	-	-
Paper 2007 [5]	-	Y	-	-	-
Paper 2014 [18]	Y	-	-	-	Y
Paper 2014 [19]	-	Y	-	Y	Y
Paper 2015 [20]	-	-	-	Y	Y
Paper 2015 [21]	Y	Y	Y	Y	Y
Paper 2017 [22]	-	Y	Y	Y	Y
Paper 2018 [23]	-	Y	Y	Y	Y
Paper 2018 [24]	-	Y	Y	-	Y
Paper 2018 [25]	-	Y	Y	Y	Y
Paper 2019 [26]	-	Y	-	Y	Y
Paper 2019 [27]	-	Y	Y	-	Y
Annual reviews [28–35]	-	Y	Y	Y	Y

PM = Particulate Matter; SPN = Solid Particle Number.

2. Definitions and Fundamentals of Gasoline Engines and Filters

2.1. Gasoline Engine Fundamentals

In the EU legislation, a positive ignition (PI) engine is the term used for the spark ignition (SI) engine, in which combustion is initiated by a localized energy supply unit from a source external to combustion. The majority of PI engines use gasoline (petrol) as fuel; other fuels, such as CNG (compressed natural gas) or liquefied petroleum gas (LPG), can also be employed in PI engines, but these are not discussed in this paper.

In conventional gasoline engines, fuel and air are mixed in the intake system and are introduced together into the cylinder. The older technology for the mixture preparation was the carburetor, later it was replaced by inlet port fuel injection (PFI) systems: initially using a single injector in the inlet manifold, and later using multipoint port injection (MPI) systems. Vehicles utilizing such mixture preparation concepts will be referred to as PFIs in this paper. When the fuel is directly injected into the cylinder the engine may be called gasoline direct injection (GDI), direct petrol injection, spark-ignited direct injection (SIDI) or fuel-stratified injection (FSI) one. In this paper, the GDI acronym will be used to refer to vehicles with such engines. Engines using some combination of PFI and GDI appeared in the Japanese and US markets in 2005 [36] and have spread since then [26,37].

GDI engines can achieve fuel economy savings compared to PFI engines. The efficiency gains mostly reside from the fact that the engine breathes and compresses air and not air and fuel mixture, the resistance to knock that allows higher compression ratio and the in-cylinder cooling of the charge. The GDI combustion systems can be distinguished depending on the concept employed to achieve charge stratification [5] to wall-guided, first-generation air-guided (or side mounted), or second-generation spray-guided (or top mounted). The three combustion modes used until recently are [8] as follows; at low load, fuel is injected during the compression stroke, the charge is stratified with an overall lean mixture. At medium load, fuel is injected during the intake stroke and the charge is homogeneous with lean mixtures. At high load, injection takes place during the intake stroke; the charge is homogeneous with stoichiometric mixtures. However, lean stratified operation has high particulate emissions [38], and needs lean-burn NO_x aftertreatment to fulfill the legislative limits [29,39]. The regeneration of the NO_x aftertreatment devices is achieved with rich operation that can increase PM emissions and fuel consumption so removing any efficiency benefits [40]. Consequently, modern GDI engines work stoichiometrically.

Gasoline engines have been using three-way catalytic converters (TWC) since the 1970s [41]. While the availability of oxygen in the exhaust gas is essential for the oxidation of CO and HC (to water and CO_2), the reduction of NO_x is inhibited by the presence of oxygen. Thus, the catalyst can simultaneously reduce all three pollutants only at stoichiometric conditions. Modern TWCs are extremely efficient with nearly complete conversion under hot engine running conditions, so most of the emissions occur during the first few tens of seconds after engine start, before catalyst light-off. Improvements in tailpipe gas emissions control from gasoline engines mostly target a faster catalyst light-off.

In the conventional PFI engine, fuel is injected into the intake port so that fuel and air flow simultaneously into the combustion chamber during the intake process, and a homogeneous air–fuel mixture is formed. On the other hand, in GDI engines, fuel is sprayed directly into the combustion chamber, which leads to incomplete fuel evaporation due to the limited time available for fuel and air mixing, resulting in localized rich combustion and particulate matter (PM) formation [42,43]. Additionally, a small amount of fuel may impinge on the piston and make direct contact with the cold cylinder walls, which may lead to diffusion combustion and subsequent PM formation [44]. Wall-wetting can induce the formation of a solid nucleation mode consisting of ash derived from metallic lubrication oil additives [45]. The PM levels are much lower for stoichiometric GDI engines. Finally, injector deposits may store fuel which is subsequently burned with sooting combustion [25,46]. Fouling of injectors has been shown to significantly increase PM emissions [47,48]. Finally, fuel composition can also influence the emission levels [49], notably the PM emissions are strongly correlated with the aromatics levels in the fuel [50]. A detailed discussion of these items has already been conducted before (Table 1) and is therefore not repeated here.

Reduction in PM emissions from GDI engines can be achieved through hardware or software improvements (including fuel injection timing, shaping and pressure) [26,51]. Another solution to reduce PM emissions from GDI engines is the use of gasoline particulate filters (GPFs) [23].

2.2. GPF Performance

It is not the intention of this paper to give an exhaustive description of the performance of GPFs, as more dedicated studies have done in the past [16,23]. Here we only provide a brief overview of some aspects of GPF performance that can be observed as one compares GDI particulate emissions over time.

The GPFs can be installed close-coupled to the engine or under the floor, with or without a TWC coating [52–54]. For a GPF, studies find lower filtration efficiency at the close-coupled position: due to the higher temperatures the flow velocity is higher [55] and a smaller soot cake is formed [56]. Catalyst coated or catalyzed or catalytic GPFs (four-way catalysts) can be used within exhaust aftertreatment systems to control gaseous emissions providing equivalent performance by replacing some or all of the catalyst volume, consequently reducing the overall volume of the aftertreatment system and enabling more compact designs [57]. Due to the continued worldwide tightening of automotive

emission standards, catalyzed GPFs can supplement the performance of the existing three-way catalytic devices [58]. GPF coating can enhance soot oxidation [59]. There is a trade-off between improved gaseous emissions conversion activity and filtration efficiency, and increased system backpressure and component cost [53,60].

2.2.1. Durability and Ash Accumulation

In EU, with Euro 5 and Euro 6, the useful vehicle life (durability) is specified at 160,000 km. California LEV III, starting in 2025, increased the durability requirements from 120,000 miles to 150,000 miles (240,000 km). In China, the durability is increased to 240,000 km starting with China 6b (2023). The durability of GPFs is affected by thermal aging, as with three-way catalytic converters, and in addition by severe thermal stress due to localized soot oxidation. Thus, the typical thermal aging of TWC is not appropriate for GPFs [61]. The proposed protocol should include thermal aging, ash loading and soot accumulation and regeneration [62]. One study that followed such protocol showed robust performance of a catalyzed GPF in the underfloor position, with filtration efficiency improving from 74% for a fresh filter to > 99% after aging equivalent to full useful life of 200,000 km (China 6b). Installed coated GPFs on vehicles have also demonstrated a good level of catalytic activity and filtration efficiency after full useful life aging and over a range of driving conditions [60,63,64].

Over their lifetime, GPFs collect inorganic ash particles derived from lubricating oil, engine wear, and corrosion of upstream exhaust pipes [65]. GPFs have to be designed to accumulate ash without a significant increase in pressure drop. Several studies have measured the ash accumulation rates, with a range of 0.1 to 0.3 mg/km, which translates to 24–72 g over the useful vehicle life of 240,000 km [66]. The accumulation of ash is associated with an increase in clean (i.e., without soot) pressure drop. However, the presence of an ash layer is also associated with an increase in filtration efficiency as the ash accumulation along the wall pores reduces soot particles from penetrating the filter wall [60,63,67]. As a consequence, and remarkably, the filtration performance of a GPF only improves with vehicle age [68]. Furthermore, the presence of ash also reduces the soot loaded pressure drop associated with deep-bed filtration. Only a small amount of ash is required to increase the filtration efficiency significantly. In one study [69] the overall filtration efficiency on a test cycle was found to increase from 75 to 90%, when the filter was loaded with 1.5 g/L of artificial ash. Mileage accumulation on vehicles equipped with GPFs shows that 3000 km can increase the filtration efficiency by 10 to 15% [68]. This justifies the deterioration factor of 1 (e.g., no deterioration) for particulate emissions in the regulations, which, however, assumes proper management of the filter to prevent damages over its useful life.

2.2.2. Regeneration

GDI engines have lower engine-out PM emissions than diesel engines, and much less soot is accumulated. Soot regeneration can occur at exhaust gas temperatures ~500 °C even with little available oxygen content in the exhaust [70]. The low temperature (200–400 °C) reaction with NO₂ and soot is limited because most of NO and NO₂ are reduced from three-way catalyst upstream of the GPF. GDI soot morphology and reactivity have also been studied and are sensitive to engine operating conditions, injection parameters, and the presence of ash [71]. Due to high temperatures associated with gasoline exhaust, the filter is expected to be passively regenerating for much of the vehicle operation. Fuel cut events associated with vehicle deceleration lead to abundant oxygen (~20% concentration) in the exhaust, which along with the high temperatures and low flow rates are sufficient to oxidize the accumulated soot [70]. Nevertheless, active regeneration strategies are also being proposed for certain conditions such as slow driving for extended durations, with low exhaust temperatures, such as those typically encountered in urban congested driving. To initiate soot oxidation, the GPF inlet temperature is elevated through engine methods (e.g., spark retard) or generating an exotherm on the three-way catalyst via air-to-fuel ratio modulation [72]. Whether initiated through passive or active means, soot oxidation is exothermic and leads to temperature rise within the GPF. The maximum temperature within the GPF depends on the soot load and inlet gas temperature and increases by ~60–100 °C per

g/L of soot load [73]. Maximum temperatures exceeding 1100 °C have been noted at very high soot loads (>5 g/L), and GPFs can withstand these high temperatures. However, the filtration efficiency might decrease due to cracks if these temperatures are exceeded.

When filter regeneration is triggered the filtration efficiency of particles >23 nm drops from approximately 90% to 60% due to the reduced soot cake [74]. The filtration efficiency for smaller particles is higher due to their higher diffusivity [51], thus higher filtration efficiencies are expected for sub-23 nm particles. Typically tailpipe emissions with the use of GPF comfortably meet the limit of 6×10^{11} particles per km even with the inclusion of particles below 23 nm [75]. However, due to the high exhaust gas temperature many particles <30 nm can be formed downstream of the GPF [56,61,74]. The nature of these particles is not clear yet: they could be renucleated semivolatile particles downstream of the thermal pretreatment unit of the particle number measurement systems or even nonvolatile particles formed between the vehicle and the dilution tunnel. A recent study showed that particle number systems at the tailpipe or the dilution tunnel can have high differences in the sub-23 nm concentrations during regeneration events [76]. Thus, more research is needed on the topic. Finally, if GPFs will include and use active regeneration systems, appropriate correction factors (K_i) that take into account the contribution of the regeneration emissions will have to be evaluated for the regulations [75].

3. Regulations

3.1. Emission Standards

In 1970, the first Emissions Directive was published under the well-known designation 70/220/EEC and focused on HC and CO emissions only [77]. Aftertreatment emission control technologies (catalysts) could not be used at the time due to the lead content in the gasoline fuel [41]. In 1984 the EC proposed that unleaded gasoline must be made available at the gas stations by 1989. This opened the door for the next step: Emission Directive 91/441/EEC (Euro 1) for passenger cars (only), which triggered the Europe-wide breakthrough of the (closed-loop) catalyst technology. Directive 93/59/EEC expanded this requirement to both passenger cars and light trucks. A fundamental prerequisite for the efficient operation of exhaust aftertreatment devices was having fuel with a very low sulfur content [78]. The next steps of Euro standards were preceded by the introduction of more stringent fuel regulations that required maximum gasoline sulfur content of 150 ppm in 2000 and 50 ppm in 2005. “Sulfur-free” diesel and gasoline fuels (≤ 10 ppm S) had to be available from 2005, and became mandatory in 2009. Euro 2 standards were introduced with Directive 94/12/EEC (passenger cars) and 96/69/EC (light commercial vehicles). Euro 3 and Euro 4 standards were introduced with Directive 98/69/EC. The Euro 5 and Euro 6 limits were introduced with Regulation 715/2007 (amended by 692/2008), and provided the automotive industry with a longer timeline to develop strategies for meeting these demanding emission limits.

Particulate matter (PM) mass emissions were first regulated as of 1992 for diesel vehicles [79]. Positive ignition vehicles were exempted from PM standards until the Euro 4 stage. Euro 5b legislation (2011) included, for the first time, a solid particle number (SPN) emission limit of 6.0×10^{11} p/km (“p” for particles from now on) for diesel (compression ignition) vehicles based on the findings of the PMP (Particles Measurement Programme) [80,81]. A SPN emission limit of 6×10^{11} p/km was introduced for GDIs with Euro 6 from September 2014 for new vehicle types and September 2015 for all vehicle models (Regulation 459/2012). However, for up to three years after these dates a particle number emission limit of 6×10^{12} p/km could be applied to Euro 6 GDI vehicles upon request of the manufacturer. This three-year phase-in was allowed in the EU to extend the period of research and development needed to meet the standard.

Table 2 summarizes the Euro standards for PI vehicles.

Table 2. European Union (EU) emission standards for positive ignition (PI) light-duty vehicles (g/km for CO, mg/km for HC, NO_x, PM, and p/km for SPN).

Euro	Class	Cycle	NM	AM	CO	HC	NO _x	CF _{NOx}	PM ¹	SPN ¹	CF _{SPN}
1	M1, M2	EDC	07/'92	01/'93	2.72	970	-	-	-	-	-
	N1-I		10/'93	10/'94	2.72	970	-	-	-	-	-
	N1-II		10/'93	10/'94	5.17	1400	-	-	-	-	-
	N1-III		10/'93	10/'94	6.90	1700	-	-	-	-	-
2	M1, M2		01/'96	01/'97	2.20	500	-	-	-	-	-
	N1-I		01/'97	10/'97	2.20	500	-	-	-	-	-
	N1-II		01/'98	10/'98	4.00	600	-	-	-	-	-
	N1-III		01/'98	10/'99	5.00	700	-	-	-	-	-
3	M1, M2	NEDC	01/'00	01/'01	2.30	200	150	-	-	-	-
	N1-I		01/'00	01/'01	2.30	200	150	-	-	-	-
	N1-II		01/'01	01/'02	4.17	250	180	-	-	-	-
	N1-III		01/'01	01/'02	5.22	290	210	-	-	-	-
4	M1, M2		01/'05	01/'06	1.00	100	80	-	-	-	-
	N1-I		01/'05	01/'06	1.00	100	80	-	-	-	-
	N1-II		01/'06	01/'07	1.81	130	100	-	-	-	-
	N1-III		01/'06	01/'07	2.27	160	110	-	-	-	-
5a	M1, M2		09/'09	09/'11	1.00	100	60	-	5.0	-	-
	N1-I		09/'09	09/'11	1.00	100	60	-	5.0	-	-
	N1-II		09/'10	01/'12	1.81	130	75	-	5.0	-	-
	N1-III, N2		09/'10	01/'12	2.27	160	82	-	5.0	-	-
5b	M1, M2		09/'11	01/'13	1.00	100	60	-	4.5	-	-
	N1-I		09/'11	01/'13	1.00	100	60	-	4.5	-	-
	N1-II		09/'11	01/'13	1.81	130	75	-	4.5	-	-
	N1-III, N2		09/'11	01/'13	2.27	160	82	-	4.5	-	-
6	M1, M2		09/'14	09/'15	1.00	100	60	-	4.5	6 × 10 ¹²	-
	N1-I		09/'14	09/'15	1.00	100	60	-	4.5	6 × 10 ¹²	-
	N1-II		09/'15	09/'16	1.81	130	75	-	4.5	6 × 10 ¹²	-
	N1-III, N2		09/'15	09/'16	2.27	160	82	-	4.5	6 × 10 ¹²	-
6c	M1, M2	WLTC	-	09/'18	1.00	100	60	-	4.5	6 × 10 ¹¹	-
	N1-I		-	09/'18	1.00	100	60	-	4.5	6 × 10 ¹¹	-
	N1-II		-	09/'19	1.81	130	75	-	4.5	6 × 10 ¹¹	-
	N1-III, N2		-	09/'19	2.27	160	82	-	4.5	6 × 10 ¹¹	-
6d-temp	M1, M2		09/'17	09/'19	1.00	100	60	2.1	4.5	6 × 10 ¹¹	1.5 ²
	N1-I		09/'17	09/'19	1.00	100	60	2.1	4.5	6 × 10 ¹¹	1.5 ²
	N1-II		09/'18	09/'20	1.81	130	75	2.1	4.5	6 × 10 ¹¹	1.5 ²
	N1-III, N2		09/'18	09/'20	2.27	160	82	2.1	4.5	6 × 10 ¹¹	1.5 ²
6d	M1, M2		01/'20	01/'21	1.00	100	60	1.43 ²	4.5	6 × 10 ¹¹	1.5 ²
	N1-I		01/'20	01/'21	1.00	100	60	1.43 ²	4.5	6 × 10 ¹¹	1.5 ²
	N1-II		01/'20	01/'22	1.81	130	75	1.43 ²	4.5	6 × 10 ¹¹	1.5 ²
	N1-III, N2		01/'20	01/'22	2.27	160	82	1.43 ²	4.5	6 × 10 ¹¹	1.5 ²

¹ Applies only to vehicles with direct injection engines. ² Kept under annual review to reflect technological progress. NM = New models (month/year); AM = All models (month/year); CF = Conformity factor; EDC = European driving cycle; NEDC = New European driving cycle; WLTC = Worldwide harmonized light vehicles test cycle.

Beginning with Euro 5 (adopted in 2009), standards are issued by Regulations, which are directly enforceable in all Member States, as opposed to Directives, which had to be transposed into each individual Member State. Note that the light-duty Euro standards use Arabic numerals, as opposed to heavy-duty Euro standards which use Roman numerals. All dates listed in the tables refer to new type approvals (TA). The EC Directives also specify a second date, usually one year later, which applies to first registration (FR—entry into service) of existing, previously type-approved vehicle models. The vehicle classes such as passenger cars (M1), small buses <5000 kg (M2), light commercial vehicles < 3500 kg (N1), or larger ones 3500–12500 kg (N2) are described in Commission Directive 2001/116/EC. For Euro 3, and later the Category N1 reference, mass classes are Class I ≤ 1305 kg, Class II 1305–1760

kg, and Class III > 1760 kg. For Euro 1 and Euro 2 the Category N1 reference mass classes were Class I ≤ 1250 kg, Class II 1250–1700 kg, and Class III > 1700 kg.

3.2. Test Cycles and Procedures

Emissions were tested over the European driving cycle (EDC) or MVEG (motor vehicle emissions group) cycle (Euro 1 and Euro 2). Effective in 2000 with the Euro 3 standard, the cycle was modified to eliminate the 40 seconds engine warm-up period before the beginning of emission sampling. This modified cold start test is referred to as the New European Driving Cycle (NEDC).

Regulation 715/2007, which established the Euro 5 and 6 engine standards, stated that the [European] Commission should “keep under review the need to revise the New European Drive Cycle.” In September 2017 the worldwide harmonized light vehicles test procedure (WLTP) and the corresponding worldwide harmonized light vehicles test cycle (WLTC) replaced the NEDC procedure for new car types (September 2018 for all new vehicles). The WLTC is based on real-world driving profiles and contains more dynamic driving conditions than the NEDC, such as higher maximum velocity and less idling time. The new testing procedure contains more differences than the speed profile alone, and these differences can be grouped in four categories [82]:

- Road load setting.
- Laboratory test set up and conditions.
- Post-processing of the test results.
- Declaration of CO₂ results.

3.3. Real-Driving Emissions (RDE)

Despite the increasing stringency of the European emission standards, a series of studies, beginning in 2011 using portable emission measurement systems (PEMS) mounted onboard vehicles [83], as well as studies utilizing remote sensing techniques [84], reported unexpectedly high real-world emissions of nitrogen oxides (NO_x) from European diesel passenger cars.

The gap between official laboratory results and the actual on-road emissions led to a revision of the emission type approval requirements in the EU. The use of PEMS and the introduction of the ‘not-to-exceed’ (NTE) regulatory concept was based on Regulation 715/2007 where it was stated that revisions may be necessary to ensure that real world emissions correspond to the levels measured at type approval. A technical working group on real-driving emissions (RDE) was set up in 2011. The work of the RDE group produced several pieces of legislation: Commission Regulation (EU) 2016/427 (first regulatory act of the RDE regulation) introduced on-road testing with PEMS to complement the laboratory Type I test for the type approval of light-duty vehicles in EU. Subsequently, Commission Regulation (EU) 2016/646 introduced the NTE concept, which equals the emission limit for the laboratory Type I test multiplied by a so-called conformity factor (CF) that takes into account the measurement uncertainty of the PEMS. Both regulations were consolidated in the WLTP EC Regulation (EU) 2017/1151, and further developed by EC Regulation (EU) 2017/1154 (the third part of the RDE Regulations), which also introduced a RDE conformity factor for the on-road test of solid particle number (SPN) emissions. The fourth part of the RDE Regulation 2018/1832 introduced on-road emissions testing as part of in-service conformity checks and slightly lowered the conformity factor for NO_x following a review [85].

The RDE test is performed during vehicle operation using a PEMS with SPN CF 1.5 from September 2017 for new models and from September 2018 for all new vehicles of types M, N1 and one year later N1-II, NI-III, and N2 vehicles. A temporary CF for NO_x was set at 2.1 and can be used upon the request of the manufacturer in Europe from September 2017 for new models and from September 2019 for all new vehicles. The final CF for NO_x for January 2020 (new models) and 2021 (all new vehicles) was originally set at 1.5 and it was reduced, following a review, to 1.43 in 2018 [85].

It should be mentioned that the NO_x gap seen with the diesel vehicles was not an issue for gasoline vehicles in Europe [83,86]. However, this was questioned by a recent study in China [87].

The main concern for gasoline vehicles was the SPN emissions: would low SPN gasoline emissions in the laboratory be reflected also in real world?

3.4. SPN-PEMS

Regulation 459/2011 required the development of test procedures to measure particle emissions from PI vehicles under real driving conditions. The Commission was tasked with developing and introducing corresponding measurement procedures at the latest within three years after the enforcement of Euro 6. To address the SPN issue, JRC evaluated the efficacy of random dynamometer cycles or on-road testing with PEMS. PEMS was the preferable option, but the technology for measuring SPN on a vehicle was not as accurate as the one in the laboratory.

In November 2012 the interest for SPN-PEMS was announced and in April 2013 the kick-off meeting of SPN-PEMS group took place. The group worked intensively and continuously until the end of 2015. In a preliminary study, a theoretical evaluation was conducted and showed that diffusion charging (DCs) based instruments are an acceptable alternative to the condensation particle counters (CPCs) of the laboratory SPN PMP equipment, with an extra measurement uncertainty of 50% [88]. At a next step (end of 2013) various prototype SPN-PEMS (DC-based) were evaluated in a chassis dynamometer lab to assess and validate the application and performance of portable SPN instrumentation. The results showed that the best performing SPN-PEMS was within 60% compared to the reference laboratory PMP system. In the second evaluation phase (which started in September 2014 and lasted until the end of that year) the best performing SPN-PEMS (DC-based, the same as in Phase I) had differences from the reference instrument at the dilution tunnel within 50% (with only a few exceptions) [89]. JRC selected the two best performing systems (one DC, and one CPC based) for the assessment of their performance through an interlaboratory comparison exercise (ILCE). The differences to the reference PMP systems were lower than 50% [90].

At this point it should be clarified that the SPN (and NO_x) CF covers only the sampling and instrument measurement uncertainty. It is well known that the vehicle SPN emissions will vary from trip to trip based on the route, traffic, driving style, altitude, ambient temperature, fuel, etc. This variability is not deemed to be included in the CF and all emission levels measured over the RDE should be below the NTE limit.

3.5. Market Surveillance

Regulation (EU) 2018/858 replaced Directive 2007/46/EC and introduced a new EU type-approval framework (from September 2020), with an effective market surveillance system to control the conformity of vehicles already in circulation (from September 2019). Member states will be able to take measures (including ordering vehicle recalls and revoking type-approval certificates) against noncompliant vehicles sold in their national markets, instead of having to wait for the type-approval authority of the country that issued the vehicles' type-approval certificate to take action.

4. Results

The first section of the results presents trends in SPN, PM (Appendix A also presents total PN), and size distributions based on an exhaustive literature search on gasoline vehicles according to the criteria described in Section 5. The emissions are plotted separately for EU, US and Asia (China, Japan, Korea) type approved vehicles, when enough data are available. At each region the respective type approval cycles are used (e.g., FTP-75 or LA92 in US, JC08 in Japan, NEDC in EU until 2017, and WLTC from 2017 in EU). It must be repeated that all results refer to vehicles over transient driving cycles and not engines operating on specific steady-state modes, to better reflect actual operation emission levels. The studies considered for each group of results are given in Appendix B. With gasoline particulate filters (GPFs) becoming popular lately, vehicles equipped with GPF are plotted separately.

4.1. PM Mass Emissions

Figure 1 shows the PM mass emissions for GDI and PFI vehicles. The PM mass limits are designated with the lines.

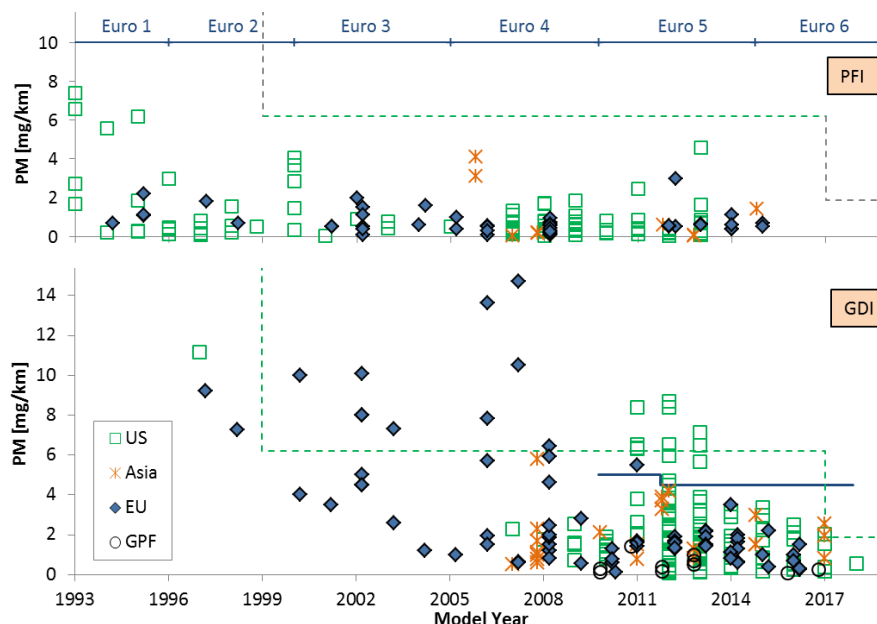


Figure 1. Particulate matter (PM) mass emissions for gasoline direct injection (GDI) (lower panel) and port fuel injection (PFI) (upper panel) vehicles. Solid line shows PM limits in EU for GDI vehicles. Dashed line shows the limits for gasoline vehicles (both GDI and PFI) in US.

The PM emissions of GDI vehicles (Figure 1, lower panel) clearly decreased over the years: from 4–14 mg/km (1996–2008) they dropped to <2 mg/km after 2009 in the EU. This does not have to do with the introduction of the PM limit, but with the introduction of the SPN limit, as it will be discussed later. The PM emissions in the US and Asia do not show a clear trend. The emissions are typically <6 mg/km in the 25 years period shown; however there are some exceptions that have emissions from 6 to 8 mg/km, probably due to the test cycles that were used (LA92 instead of FTP).

The PM mass emissions of PFI vehicles (Figure 1, upper panel) are low and typically <2 mg/km for all regions. Higher emissions were measured for older vehicles (pre-2000) [91,92]. Some aggressive cycles, such as the Common Artemis Driving cycles (CADC), gave also emission levels around 10 mg/km (such cases not plotted in the figure). PM limits in the US were tightened from 6.2 mg/km (1999–2017, Tier 2) to 1.8 mg/km (from 2017, Tier 3). In California, a further reduction to 0.62 mg/km is phased-in starting 2025. Interestingly there are no PM data for US MY after 2013, probably due to the increased research interest on GDIs and their high market share. In China, a PM limit of 4.5 mg/km applies to GDI vehicles only, starting with China 5. The limit continues through China 6a (2020), and further tightens to 3 mg/km with China 6b (2023).

4.2. Chemical Composition

PM emissions from PFI vehicles are generally very low to allow for an accurate characterization of their chemical composition. Under hot engine conditions, most of the emitted PM is elemental and organic carbon [93,94]. Studies in the early 2000s, where the PM levels were ~0.5 mg/km, found elemental carbon (EC) percentages of less than 30% over NEDC cycles [95,96]. Engine studies found even lower EC fractions (<5%) [97,98]. EC can be higher under transient engine operation or at low ambient temperatures [99]. A US study found percentages of 30 to 65% (black carbon) over FTP cycles [100]. Another study reported that the EC fraction increased from 40% to 75% when the temperature decreased from 22 °C to −1 °C [101].

Most of the PM emitted by GDI vehicles is found to be EC (on average 80%; range: 45–95%). The high EC content is consistent since the introduction of GDIs in the markets: from early 2000s [93,95] to 2010s [94,100,102–109]. The vehicle to vehicle variability masks any influence of test cycle, position of the injectors (side or top), or fuel injection strategy (stratified or stoichiometric) on the relative contribution of EC to PM. Engine studies have also shown high EC percentages (70–93%) [98,110], but other studies have found much lower percentages (15–35%) [111,112]. With dedicated engine studies it was also shown that idle or fuel cut conditions can increase the volatile part [110] and that the EC fraction is higher under lean stratified conditions (80%) compared to stoichiometric (60%) [113].

Other PM components are S, Fe, Ca, P, and Na [97,98,110,114]. Ash in the engine-out PM is mostly derived from lube oil additives [98,110]. Detachment of some support materials from the catalytic converter also contributes [110]. The ash that was found in a GPF after 240,000 km of driving consisted of approximately 50% engine oil additive components (Ca, P, S, and Zn), 20% corrosion material (mainly Fe_2O_3), and 30% catalytic washcoat from the three-way catalyst [65]. Polycyclic aromatic hydrocarbons (PAHs) can also be found in significant concentrations when there is no GPF [99–101,115–118]. PAHs are a concern because of their carcinogenicity and mutagenicity [119]. GPFs can reduce PAHs >60% [115,120].

4.3. SPN Emissions

Figure 2 shows the SPN trends for GDI and PFI vehicles. EU has SPN limits only for GDI vehicles (shown with solid line), while the China 6 limits will apply to both GDI and PFI vehicles (starting 2020, nationwide). Note that the SPN measurement protocol was established with the introduction of SPN limits for diesel vehicles (2011), thus measurements at earlier years were not always fully compliant with the protocol. Only measurements that used a thermal conditioning stage of at least 250 °C were considered in the results. Total PN (including volatiles and particles <23 nm) are discussed in Appendix A.

The decreasing trend of GDI emissions with time is clear (Figure 2, lower panel). The emissions were $>10^{13}$ p/km when GDIs were first widely introduced in the market [9]. A further reduction was noticed [121] with the introduction of stratified and spray-guided GDIs in 2006 [14]. The laboratory SPN limit of 6×10^{11} p/km for the NEDC was achieved with a margin (4.4×10^{11} p/km) in 2011 only with in-cylinder measures [42,122] (shown as point in MY 2010.3). In 2014 it was reported that the limit could also be achieved with a prototype vehicle on the road at moderate driving [123]. However, with aggressive driving (many high load accelerations) the SPN emissions were higher than the limit. The first on-road evaluations with Euro 5 and Euro 6b vehicles showed that the emissions remained below the 6×10^{12} p/km limit [124,125] and they were by a factor of two higher compared to the laboratory results [126]. Recently a commercial non-GPF GDI vehicle achieved emissions much lower than the SPN limit under normal driving conditions [127]. Non-GPF GDIs may achieve the RDE limit under mild conditions of use [128], although with high aromatic content fuel, severe conditions of testing, and low ambient temperature, the same vehicle may emit more than the RDE limit [129]. Another concern is the increase of emissions due to deterioration of the injectors [130].

The first GPF investigations were reported in 2011 [55]. While this was preceded by extensive experience with DPFs, the differences in operating conditions for the GPFs were highlighted: lower engine out SPN levels, higher exhaust gas temperatures, and lower oxygen concentration. Investigations included GPF porosity, pore size, cell structure, dimensions, position (closed-coupled or underfloor), and the presence and amount of catalytic coating [23,58,60]. Numerous researchers have since demonstrated the performance of GPFs, quantifying key metrics such as filtration efficiency, pressure drop, and gas conversion (for catalyzed filters) [23]. Several studies have also shown that the filtration efficiency only improves with mileage accumulation [60,63,65,131]. This is due to the accumulation of an ash layer on the channel walls, which prevents further penetration of soot particles [68]. The first commercial GPF equipped vehicle was introduced in the market in 2014 and was tested for particle emissions in 2016 [132]. The emissions were $<10^{11}$ p/km under all driving conditions. Analysis of

type approval RDE data from September 2017 of >340 vehicles from the database of the European Automobile Manufacturers Association (ACEA) [133] and the Japan Automobile Manufacturers Association (JAMA) [134] gave a mean of 1.3×10^{11} p/km and a median of 9.5×10^{10} p/km.

Figure 2 (upper panel) plots the emissions of PFI vehicles. There is no clear trend on the emissions over the last 25 years. Most of the results range between 1×10^{11} p/km to 3×10^{12} p/km. What is rather surprising is that 31% of the EU sample have emissions $>6 \times 10^{11}$ p/km. This percentage remains similar even if only vehicles after 2011 are considered (where a SPN limit was applicable to diesel vehicles). Recent on road tests with PFI vehicles (including hybrids) showed that the emissions are $<1 \times 10^{12}$ p/km [135–137].

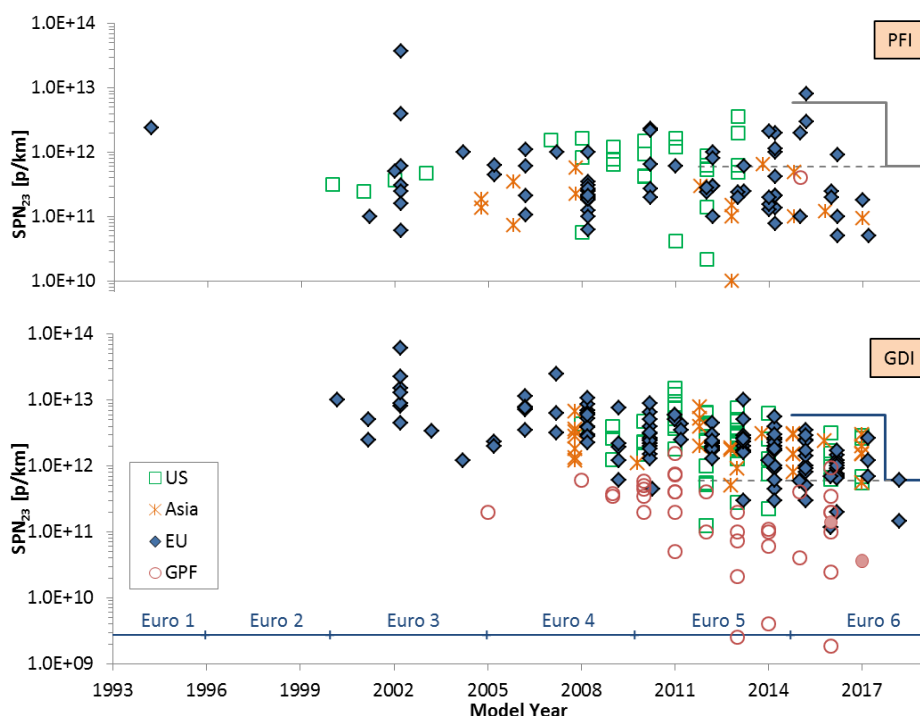


Figure 2. Solid particle number (SPN) emissions for gasoline direct injection (GDI) (lower panel) and port fuel injection (PFI) (upper panel) vehicles. Dashed line shows SPN limit for diesel vehicles (only EU). Solid line shows SPN limits for GDI vehicles (only EU). The solid circles are commercial GPF equipped vehicles (open circles are GPF retrofitted vehicles).

4.4. Size Distributions

Figure 3 summarizes the GMDs (geometric mean diameters) of GDIs and PFIs with or without thermal pretreatment of the sampled particles. Each point is the reported mean of a vehicle over the type approval cycle, typically measured with size spectrometers such as EEPS (Engine Exhaust Particle Sizer), DMS (Differential Mobility Spectrometer), and ELPI (Electrical Low Pressure Impactor) [79]. The “solid” size distributions were measured downstream of a thermodenuder or a catalytic stripper heated at temperatures >250 °C [79]. The pre-2005 GDI vehicles have GMDs of solid particle size distributions between 60 and 90 nm and the PFIs between 45 and 90 nm. The post-2008 vehicles have GMDs of solid particle size distributions between 25 and 70 nm and the PFIs between 20 and 60 nm. The shift downwards is probably linked with improved combustion and charge mixing. Boosted engines have been shown to produce particles with smaller sizes [24]. While at the same time the particle number concentrations were reduced, the smaller sizes below approximately 30 nm have higher deposition fraction in the lungs [138]. The GMDs of total particles size distributions are slightly wider extending to smaller sizes because the (additional) volatile nucleation mode decreases the GMD. The calculated average geometric standard deviation (GSD) of GDIs was 1.9 and of PFIs 2.0, in good agreement with those reported by others [139].

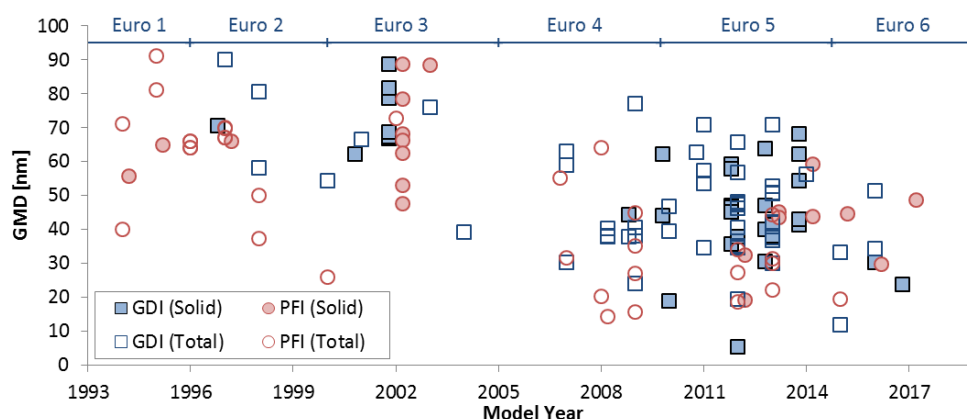


Figure 3. Geometric mean diameter (GMD) of solid and total (solid + volatiles) size distributions for gasoline direct injection (GDI) and port fuel injection (PFI) vehicles. Results from all regions (EU, US, and Asia).

4.5. Transmission Electron Microscopy (TEM) Studies

Transmission electron microscopy (TEM) can be used to provide information regarding the shape and structure of the particles collected. Energy dispersive spectrometry (EDS) can provide their elemental composition. The first TEM image of an agglomerated soot particle of a GDI vehicle at constant speed was shown in 2004 [140]. The primary particles diameter was 28 ± 3 nm. Later studies with GDI engines found that primary particles were distributed over a range of 7 to 60 nm in diameter [71,111,141]. For early fuel injection strategy, the primary particle diameter was between 20 and 25 nm with graphitic structure. For a fuel injection strategy, which produced low particle number concentration, there were many single solid sub-25 nm particles and fractal like agglomerates with primary particle size between 10 and 15 nm with more amorphous structure. Later it was shown that the primary particle size correlated with the aggregate size [142]. Other studies found primary particle sizes between 20 to 30 nm [143–148]. Recent studies with GDI vehicles, in addition to soot particles, also found oil derived ash particles in the size range of 10–25 nm [149,150], emitted, e.g., during engine braking [151].

4.6. Low Ambient Temperature

Figure 4 plots the PM mass emission of GDI (diamonds) and PFI (circles) vehicles at ambient temperatures of ~ 23 °C (solid symbols) and -7 °C (open symbols) tested in the laboratory under type approval cycles. In all cases there is an increase of the PM mass at lower temperature. For PFIs, the increase was small in some instances (+1 mg/km), while in others it was quite high, leading to PM emissions in the 8 to 12 mg/km range. For GDIs the -7 °C tests resulted in PM mass > 5 mg/km, with one vehicle at 14 mg/km. Only the GDI retrofitted with a GPF kept the PM mass below the 2 mg/km level.

Figure 5 plots the SPN emissions of PFIs (triangles, upper panel) and GDIs (diamonds or circles when equipped with GPF, lower panel) at ambient temperatures of around 23 °C (solid symbols) and -7 °C (open symbols). Similar to mass emissions described above, the SPN emissions consistently increased when the test was conducted at -7 °C. For emission levels above 1×10^{12} p/km at 23 °C the emissions on average doubled at -7 °C [152]; however, for lower emission levels they increased up to 3–4 times. While none of the Euro 6 vehicles exceeded the 6×10^{12} p/km limit, all exceeded 6×10^{11} p/km at -7 °C. For those GDIs that were equipped with GPF, their tailpipe emissions did not significantly increase at the -7 °C tests and they remained below the 6×10^{11} p/km limit (MY after 2011).

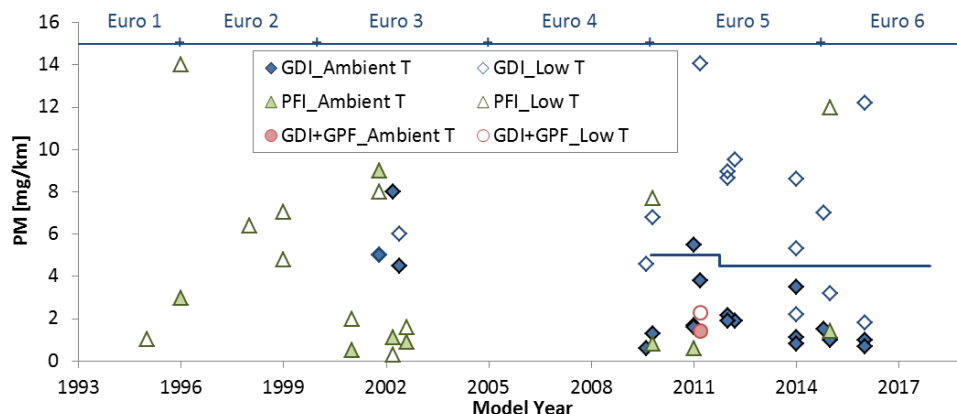


Figure 4. Particulate matter (PM) mass emissions for gasoline direct injection (GDI) (lower panel) and port fuel injection (PFI) (upper panel) vehicles at different ambient temperatures. “Ambient T” refers to approximately 23 °C. “Low T” refers to −7 °C. Solid line shows PM limits for GDI vehicles at 23 °C (only EU).

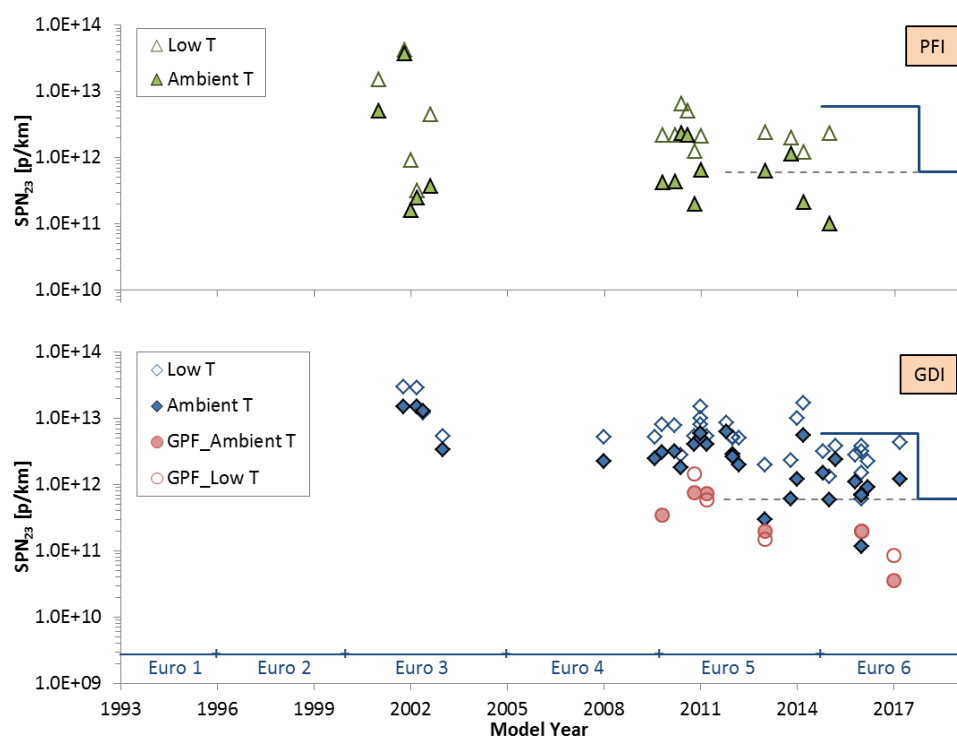


Figure 5. Solid particle number (SPN) emissions for gasoline direct injection (GDI) (lower panel) and port fuel injection (PFI) (upper panel) vehicles at different ambient temperatures. “Ambient T” refers to approximately 23 °C. “Low T” refers to −7 °C. Dashed line shows SPN limit for diesel vehicles at 23 °C (only EU). Solid line shows SPN limits for GDI vehicles at 23 °C (only EU).

Currently, gasoline vehicles have limits for CO and HCs at −7 °C. Discussions are ongoing to introduce a low temperature procedure in the regulations. The results of the two figures can support in deciding future limits.

4.7. Sub-23 nm Fraction

Figure 6 summarizes the results of studies that measured (solid) particles sized below 23 nm, which is currently the lower size limit defined in the EU regulation. Additionally, based on the size distributions of solid particles shown in Figure 3, the fraction of particles below 23 nm was estimated assuming a lognormal distribution and was added in Figure 6. The assumption of monomodal size

distributions is not necessarily true and underestimates the cases with a separate nucleation mode. Until early 2000s, there are only estimations of the particle number fraction below 23 nm. Due to the large GMD, this fraction is found to be below 15% of the total number. The estimated percentage after 2009 raises up to 45% for both GDIs and PFIs (one 2012 GDI estimated at close to 100%). The measured percentage for GDIs is in the same range with a few measurements in the 45 to 60% range. However, for PFIs the measured percentage is up to 75% (95% in one case).

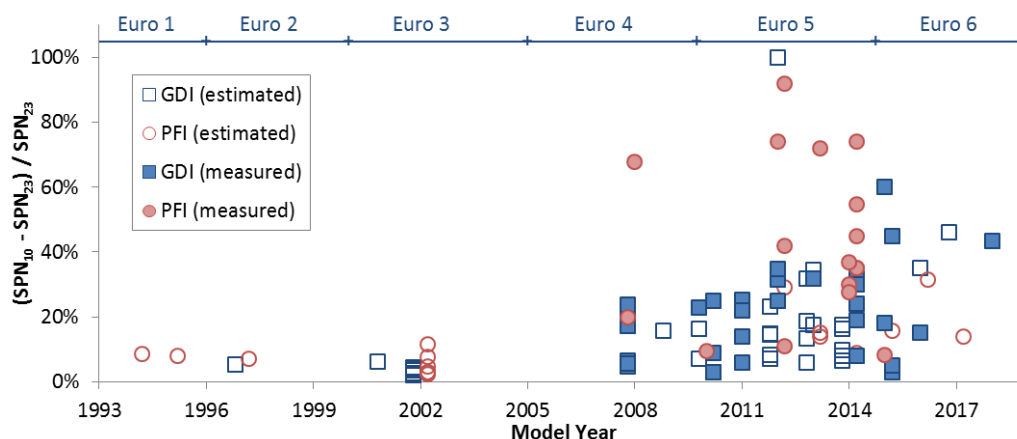


Figure 6. Additional particles below 23 nm. Measured: Based on 23 nm and 10 nm condensation particle counters (CPCs) downstream of a thermal pretreatment unit. Estimated: Based on solid size distributions (Figure 3). No corrections for particle losses in the measurement systems.

Figure 7 plots the ratio of particles above over below 23 nm as a function of the emission levels following the regulatory method (SPN_{23}). Generally, there is a tendency of a higher percentage with lower emissions. This is an indication that for high concentrations coagulation takes place and increases the GMD [153]. In addition, the relative contribution of lubricant increases as soot from combustion decreases. The percentage can be $>30\%$ for emission levels $<1 \times 10^{12}$ p/km.

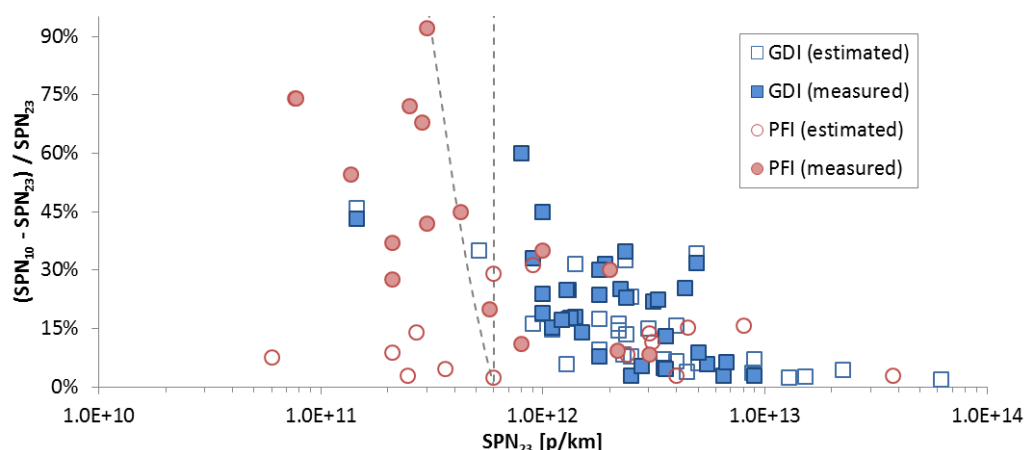


Figure 7. Additional particles below 23 nm as a function of the emission levels of particles >23 nm. Measured: Based on 23 nm and 10 nm condensation particle counters (CPCs) downstream of a thermal pretreatment unit. Estimated: Based on solid particle size distributions (Figure 3). No corrections for particle losses. Dashed lines show the 6×10^{11} p/km limit for 23 nm (vertical) and 10 nm (diagonal).

Keeping the same limit 6×10^{11} p/km but changing the lower size from 23 nm to 10 nm would result in 2 out of 15 measured cases failing the new methodology. Thus, the current SPN methodology can still identify most, but not all, high emitting gasoline vehicles. The number of cases would increase with a lower limit, for example 3×10^{11} p/km. In any case, when the fraction of total emissions not

measured with the current methodology is significant, decreasing the threshold below 23 nm would provide a more comprehensive control of future gasoline vehicle emissions.

5. Materials and Methods

The analysis and the graphs of this study are based on a literature survey on particle emissions of gasoline vehicles using relevant terms (gasoline, direct injection, port fuel, multipoint injection, GDI, DISI, MPI, PFI, particulate emissions, particle number, size distribution, and TEM) and in addition scanning the references cited therein. The vehicles had to be type approved according to the Euro 1 emission standards or the model year (MY) had to be 1993 or later.

Only vehicle studies were considered. Only results with market or reference fuels with ethanol content up to 10% were used. Studies with addition of additives, oils etc. in the fuel were not included. For the solid particle number (SPN) analysis (total or GMD) only studies following the regulated procedure, or using thermodenuder, or catalytic stripper with temperature $>250\text{ }^{\circ}\text{C}$ were analyzed. For total particle size distributions only studies without any thermal pretreatment were accepted (i.e., dilution with air at ambient temperature). The GMD was estimated by digitally replotting the size distributions from the relevant studies. Similarly, for the particle number emissions when the studies did not report the values in the text, the values were extracted digitally from the plotted figures so some uncertainty based on the resolution can be expected.

When enough data was available the results were split in the EU, US and Asia type approved vehicles. When the model year (MY) of the vehicle was not given it was assumed to be 1–2 years before the specific publication or based on the standard it followed. For the graphs the MYs were shifted ± 0.2 years randomly to improve the readability.

The studies that were used in each figure are given in the Appendix B.

6. Conclusions

Emissions of gaseous pollutants from gasoline vehicles have been controlled since the 1970s. Significant reductions were achieved with the introduction of three-way catalysts in the 1990s. Particulate matter emissions were not considered an issue, because they were orders of magnitude lower than from diesel vehicles. The picture changed when particulate filters were installed on diesel vehicles and gasoline direct injection (GDI) vehicles were introduced in the market. The introduction of a solid particle number limit for the GDIs resulted in a significant decrease of their particulate emissions in terms of mass, solid, and total particle number concentrations. The new additional legislation requirement of achieving the same levels also on the road under real driving conditions resulted in installation of particulate filters for GDIs in EU models. Data included here showed that in some cases, emissions levels from PFI vehicles can exceed those from GDI vehicles. The low ambient temperature conditions further increase the emissions. The measured particle concentration below 23 nm is in many cases $>50\%$ (without correcting for particle losses). These results suggest that for post Euro 6 regulations, assuming that PFIs still have an important market share, they should be included in the next regulatory step, the lower limit of 23 nm should be decreased to 10 nm and low ambient temperature tests for SPN should be included.

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Disclaimer: The opinions expressed in this manuscript are those of the authors and should in no way be considered to represent an official opinion of the European Commission. Mention of trade names or commercial products does not constitute endorsement or recommendation by the authors or the European Commission.

Appendix A

Figure A1 plots the total PN emissions for GDI (lower panel) and PFI (upper panel) vehicles. Different instruments were used (such as CPCs or EEPs) with different lower cut-off sizes (typically 2.5–6 nm). The majority of the measurements was conducted from the full dilution tunnel (CVS) where the dilution factor changes over a test. As with SPN, a decreasing trend can be seen for GDIs, but no trend for PFIs. As discussed elsewhere, the reproducibility of total PN measurements from the CVS is not good due to the variant dilution factor, the different flow rates employed at CVS, and the different residence times. In addition, the volatiles desorption from the transfer tubes to the CVS during high speed or aggressive cycles can result in unrealistically high values [107]. The opinion of the authors is that total PN should be conducted from the tailpipe under well-defined sampling conditions [154–156].

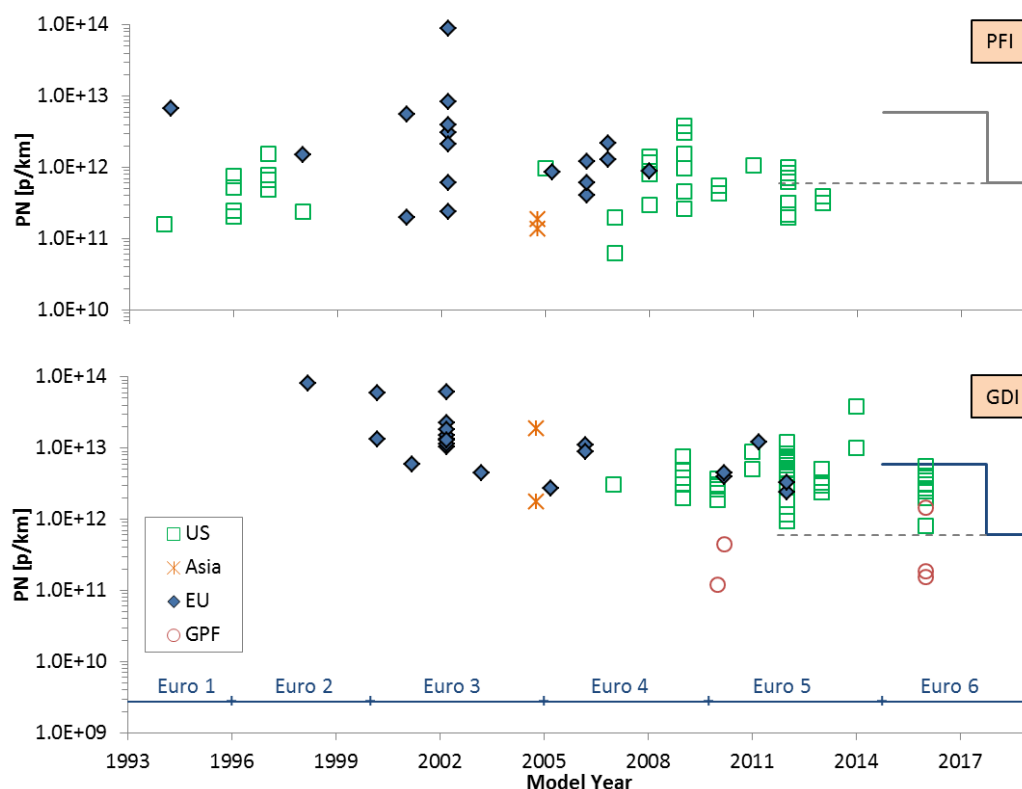


Figure A1. Total particle number (PN) emissions for gasoline direct injection (GDI) (lower panel) and port fuel injection (PFI) (upper panel) vehicles. Dashed line shows solid PN (SPN) limit for diesel vehicles (only EU). Solid line shows SPN limits for GDI vehicles (only EU).

Appendix B

The following studies were considered in the figures of the main text.

Figure 1 (upper panel): PM PFI: [42,91,93–96,100,101,121,157–194]

Figure 1 (lower panel): PM GDI: [37,42,51,68,74,93–95,100,102–106,108,109,120,121,132,157–163,165–167,169,170,173,175–179,181,184,186,188–190,194–225]

Figure 2 (upper panel): SPN PFI: [42,51,75,93,96,121,127,158–165,168–183,194,224,226–242]

Figure 2 (lower panel): SPN GDI: [18,42,53,55,56,58,60,61,63,68,74,75,93,103–105,109,114,115,120–124,126,127,132,137,150,158–165,169–171,173,175–181,194,197–203,205–209,211,214,218,222–226,228,230–235,237–241,243–258]

Figure 3: GMD (Solid): [18,75,89,105,127,137,150,157,158,168,205,223,229,230,245,247,248,259–262]

Figure 3: GMD (Total): [56,94,95,102,104,109,121,140,165–168,170,172,175,176,178,181,184,189,195,198,204,210,212,213,231,236,237,254,263–271]

Figure 4: Low temperature PM: [99,158,163,165,169,176,192,196,211,224,225]

Figure 5: Low temperature SPN: [60,75,158,163,165,169,176,196,201,211,224–226,234,240,257]

Figure 6: Sub-23 nm (measured): [18,75,127,163,177,223,247,248]

Figure 6: Sub-23 nm (estimated): As Figure 3: GMD (Solid).

Figure 7: As Figure 6.

Figure A1: Total PN [93,95,104,105,108,109,150,158,161–163,170,195,204,212,226,248,271–274]

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