

Overview of Vehicle Exhaust Particle Number Regulations

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Abstract: Vehicle emissions are a significant source of air pollution in cities. Particulate matter (PM) is a pollutant with adverse health effects. Regulations worldwide determine the PM exhaust emissions of vehicles by gravimetric quantification of the mass deposited on a filter over a test cycle. The introduction of particulate filters as vehicle exhaust gas aftertreatment devices led to low PM emissions. A particle number methodology (counting solid particles > 23 nm), complementary to the PM mass measurement, was developed by the PMP (Particle Measurement Programme) group of the GRPE (Working Party on Pollution and Energy) of the UNECE (United Nations Economic Commission for Europe) during the first decade of the 21st century. The methodology was then introduced in the EU (European Union) regulations for light-duty (2011), heavy-duty (2013), and non-road mobile machinery (2019). In parallel, during the last 15 years, UN (United Nations) regulations and GTRs (Global Technical Regulations) including this methodology were also developed. To address the on-road emissions, the EU introduced RDE (real-driving emissions) testing with PEMS (portable emissions measurement systems) in 2017. Other countries (e.g., China, India) have also started adopting the number methodology. The PMP group recently improved the current laboratory and on-board methodologies and also extended them to a lower particle size (counting solid particles > 10 nm). Due to the rapid evolution of the vehicle exhaust particle number regulations and the lack of a summary in the literature, this paper gives an overview of current and near future regulations. Emphasis is given on the technical specifications and the changes that have taken place over the years.

Keywords: air pollution; vehicle exhaust emissions; particulate matter; particle number; catalytic stripper; Particle Measurement Programme (PMP); portable emissions measurement system (PEMS); emission regulations; real-driving emissions (RDE)



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1. Introduction

Air pollution affects the health of millions of people worldwide [1]. The importance of air pollution is even higher for cities, where, according to statistics, >70% of the population lives (>50% for Asia) [2,3]. Many countries have introduced policies to mitigate the increasing anthropogenic contribution to air pollution. Airborne particulate matter (PM) is one of the most relevant pollutants to human health, with both short- and long-term exposure linked to increased mortality [4,5]. PM can be emitted directly by the sources (primary) or formed in the atmosphere by gaseous precursors (secondary) [6,7]. Globally, between 2010 and 2016, 55.3% of the world's population was exposed to increased levels of PM_{2.5} [8]. It was estimated that in 2019, exposure to PM_{2.5} caused 7% of total global mortality and accounted for about 4.1 million deaths [4], largely in developing countries such as China and India. A review that summarised European data from 2000 to 2012 showed that transport was a major contributor to urban air pollution, with 27% of PM originating from road transport in the cities [9] (the percentage was 20% for overall Europe). The stricter emission standards (the so-called Euro standards), along with fuel improvements and traffic management, decreased both primary and secondary aerosol contributions from vehicles [10]. In Europe, for example, the (total) particulate emissions from road transport

decreased significantly over the years even though the transport activity increased [11]. In 2018 the road transport contribution to primary PM (overall Europe) was 11% [11]. During the COVID-19 lockdowns, the decreases in PM in cities were modest [12–15], confirming that in terms of mass, other sources are more important than transport (e.g., residential heating, manufacturing industry). Still, the reductions in urban sites were much higher compared to rural sites, indicating that traffic still contributes in cities. However, a study concluded that further controls on traffic emissions will have limited benefit in reducing the concentration of fine particles, except in countries where the use of diesel particle filters is not mandatory (e.g., outside the EU) [8,16]. Indeed, where policies and strategies are not implemented, air quality continues to worsen due to the increasing number of vehicles and power plants [17,18].

Controlling the PM mass does not necessarily address the problem of ultrafine particles concentration in the air. A study confirmed that the sources dominating the particle number emissions are different to those dominating the mass emissions [19,20]. The particle number concentration is highly affected by road transport and the mass by aged and transported aerosol [19]. In 2010, the major global particle number source was road transport (40%) [21]. In Europe, the percentage was 60%, ranging from 32% to 97% [22]. Another study with data from 2013 to 2016 showed that road transport was the main contributor in all cities examined, with a contribution up to 94% (annual average) [23]. The recent WHO (World Health Organization) Guidelines recommended the measurement of ambient ultrafine particles at monitoring stations and suggested that low particle number counts should be considered less than $1000 \text{ \#}/\text{cm}^3$ (as a 24-h mean), while typical levels observed in urban background areas typically exceed $10,000 \text{ \#}/\text{cm}^3$ [24]. The non-volatile part is typically less than one-third [25].

The methodology to assess the emissions of a vehicle is traditionally based on the weight increase in a filter sampling diluted exhaust over a test [26]. The vehicle is fixed on a chassis dynamometer, and the driver follows a pre-defined speed profile (test cycle). The exhaust is diluted in a dilution tunnel, where various analysers determine the pollutants. A part of the diluted exhaust gas is extracted and passes through a filter. The quantity collected during the test determines the PM mass emissions. With the introduction of particulate filters at the exhaust aftertreatment of diesel vehicles, the collected mass on the filter reached values close to the background levels. For this reason, the PMP (Particle Measurement Programme) group was tasked by the UNECE (United Nations Economic Commission for Europe) with developing a complementary method (to PM mass) with better sensitivity at low emission levels. The JRC (Joint Research Centre) of the European Commission actively participated in the PMP, providing data and drafting the relevant regulations since almost the beginning. The candidate method that was finally selected was based on counting solid particles with diameter $> 23 \text{ nm}$ [27]. The choice of not measuring smaller particles, which are very sensitive to the sampling conditions, was based on technical reasons (better accuracy, repeatability, acceptable cost) rather than on health reasons. Additionally, the limits were selected on the basis of what was achievable with the best available technology (wall flow particulate filters) and not on health-related considerations. The reason for this choice was based on the fact that, while a clear relationship between adverse effects and exposure to PM had been already demonstrated, there was a lack of robust epidemiological studies on the health effects of ultrafine particles levels [28]. The solid particle number (SPN) methodology was initially introduced in the EU (European Union) regulation in 2011 for light-duty diesel vehicles (Euro 5b). Since then, it has been extended to other sectors and regulations (EU and Global). Other than the EU, many other countries have introduced SPN in their regulations. For example, China (China 5 for light-duty diesel vehicles, China 6/VI all), India (BS VI all since 2020), and Korea have SPN limits. At the time of writing, in addition to the EU, only China has on-road SPN limits, and India will introduce them in 2023. Interestingly, even though only solid particles are limited, a review (years 1994–2019) showed that for gasoline direct injection vehicles where the SPN emissions decreased over the years, the total particles (i.e., solids and volatile

primary particles) also decreased. On the other hand, for port fuel injection vehicles, where no SPN limit applies, the solid particle emissions remained at the same levels or even increased, as well as the total particles [29]. Similarly, DPFs were found to be very effective in reducing secondary aerosol formation [30]. It should be emphasised, though, that total particles are more sensitive than solid particles, and they do not necessarily correlate for all cases (vehicles, fuels, test cycles etc.). Consequently, large differences between the two can sometimes be found [31].

Since the latest reviews in 2015 and 2018 [32,33], corrections and improvements to the methodologies used to measure particle numbers from motor vehicles took place, sometimes with important differences. The aim of this paper is to update on the status of current particle number regulations and, most importantly, to summarise the technical specifications, differences, improvements, and lower detection sizes (from 23 nm to 10 nm), which are not so easy to identify. To our knowledge, this is the first study that summarises all particle number regulations.

2. Regulatory Background

This section will give the background and regulatory context of the group and committees that were responsible for preparing the particle number technical specifications and adopting the relative regulations.

2.1. World Forum for Harmonization of Vehicle Regulations (WP.29)

The circulation of vehicles in a country requires the approval of the vehicle type and/or its components. Different national regulations and approval procedures mean additional time and costs for the manufacturers due to additional designs, tests, and approvals. Thus, it is highly desirable to harmonise the technical requirements and regulations for the vehicle type approval at an international or even a global level. The main organisation where such effort is taking place is the World Forum for Harmonization of Vehicle Regulations (WP.29) [34] under the United Nations Economic Commission for Europe (UNECE) [35]. There are six subsidiary bodies, also known as GRs (Groups of Rapporteurs) (Figure 1). The one relevant to vehicle emissions is the Working Party on Pollution and Energy (GRPE) [36]. Any United Nations (UN) member country and any of their regional economic integration organisations may participate in the activities of the World Forum and may become a contracting party to the Agreements (which will be discussed below). Governmental and non-governmental organisations (NGOs) may also participate in a consultative capacity in WP.29 or in its subsidiary working groups. The WP.29 convenes three times a year (March, June, and November). The sessions of each subsidiary body are usually held twice a year. The work of the World Forum is transparent: all agendas, working documents and reports are openly accessible on the Internet website of the World Forum.

Three UN Agreements, adopted in 1958, 1997, and 1998, provide the legal framework allowing contracting parties to establish internationally harmonised regulatory instruments concerning the certification of motor vehicles and their equipment and parts, as well as rules for technical inspections of vehicles in use [37].

1. UN 1958 Agreement [38];
2. UN 1998 Agreement [39];
3. UN 1997 Agreement [40].

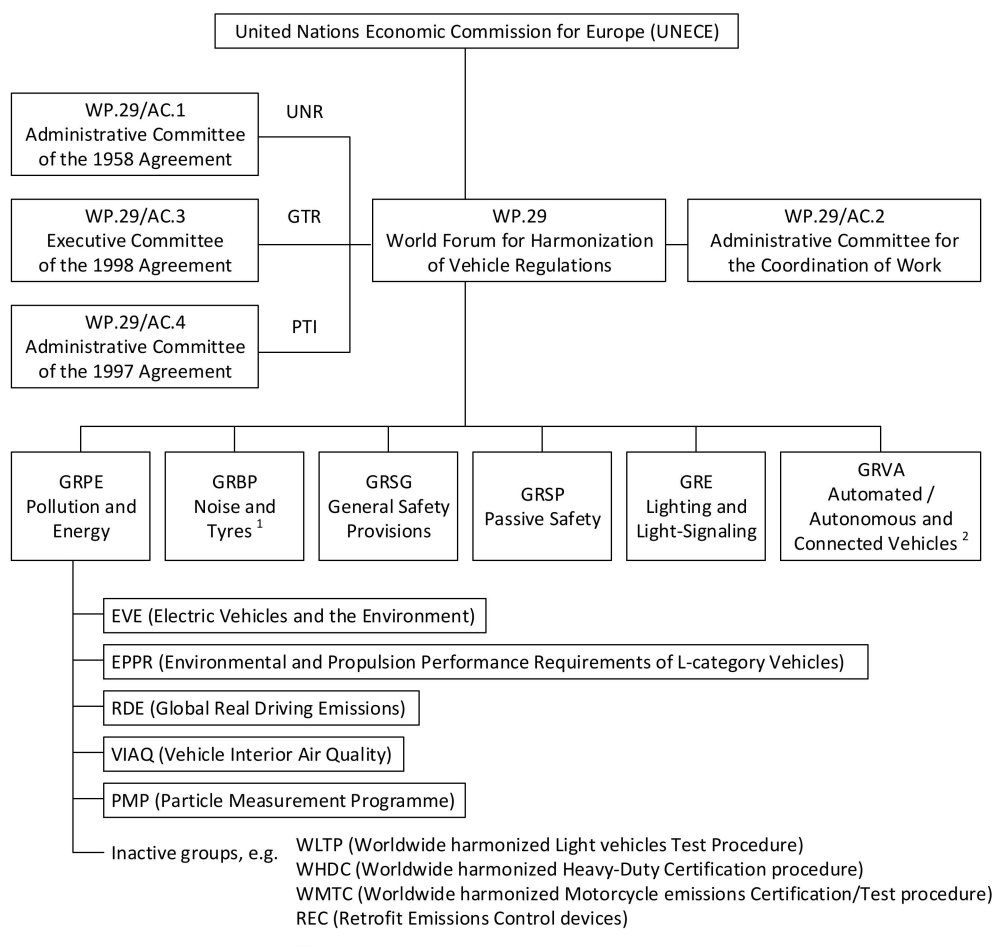


Figure 1. Organisation of the World Forum. ¹ Former Working Party on Brakes (GRB); ² Former Working Party on Brakes and Running Gear (GRRF). UNR = United Nations Regulation; GTR = Global Technical Regulation; PTI = Periodic Technical Inspection.

2.1.1. UN 1958 Agreement

The UN 1958 Agreement provides the legal and administrative framework for developing harmonised technical UN Regulations on uniform performance requirements, for granting type-approvals and documentation, for the conformity of production, and for the assessment of technical services. The basic main principles of the 1958 Agreement are: (i) the mutual recognition of the type-approvals granted by contracting parties, and; (ii) a contracting party can choose which, if any, of the UN Regulations annexed to the Agreement it would like to apply. Furthermore, even when a contracting party applies a UN Regulation, it may keep its own alternative national/regional legislation. The regulations were formerly called “UNECE Regulations” or, less formally, “ECE Regulations” in reference to the Economic Commission for Europe (ECE). However, since many non-European countries are now contracting parties to the 1958 Agreement, the regulations are officially entitled “UN Regulations” or sometimes abbreviated as “UNRs”. Some important regulations for vehicle exhaust emissions are:

- UN Regulation 40 for motorcycles. There is no particle number measurement requirement.
- UN Regulation 47 for mopeds. There is no particle number measurement requirement.
- UN Regulation 49 for heavy-duty engines.
- UN Regulation 83 for light-duty vehicles.
- UN Regulation 96 for non-road mobile machinery.
- UN Regulation 132 for retrofitting.
- UN Regulation 154 for light-duty vehicles following the worldwide harmonised light vehicles test procedure (WLTP) [41].

The oldest of the above are UNR 40 (entry into force 1979) and UNR 47 (entry into force 1981), and the latest is UNR 154 (entry into force 2020). At the time of writing, 159 UN Regulations exist, which can be found at the relative website [42].

2.1.2. UN 1998 Agreement

The UN 1998 Agreement (or Global Agreement) stipulates that contracting parties establish, by consensus vote, UN Global Technical Regulations (UN GTRs) in a Global Registry on globally harmonised performance requirements and test procedures. Each UN GTR contains extensive notes on its purpose and development. The technical rationale, the research sources used, the cost and benefit considerations, and the references to data that were consulted are recorded. GTRs are not binding to contracting parties, and they should apply their national rulemaking procedures if and when transposing UN GTRs into national legislation. At the time of writing, 21 UN GTRs exist, which can be found at the relative websites [43]. The most important for vehicle exhaust regulations are:

- GTR 2 for L-category vehicles (test cycle: worldwide harmonised motorcycle test cycle—WMTC). There is no particle number measurement description.
- GTR 4 for heavy-duty engines (worldwide harmonised heavy-duty certification procedure—WHDC) (test cycles: worldwide harmonised transient cycle—WHTC; worldwide harmonised stationary cycle—WHSC). There is no particle number measurement description.
- GTR 11 for non-road mobile machinery (NRMM) (test cycles: non-road transient cycle—NRTC; non-road stationary cycle—NRSC). There is no particle number measurement description.
- GTR 15 for light-duty vehicles (worldwide harmonised light vehicles test procedure—WLTP) (test cycle: worldwide harmonised light vehicles test cycle—WLTC).

2.1.3. UN 1997 Agreement

The UN 1997 Agreement allows contracting parties to establish UN Rules for the periodic technical inspections (PTI) of vehicles in use. The contracting parties reciprocally recognise the international inspection certificates that were granted according to the UN Rules annexed to the Agreement. The topic will not be discussed in more detail here.

2.1.4. Differences between UN 1958 and 1998 Agreements

While both the 1958 and 1998 Agreements prepare regulations on similar technical topics, the UN Regulations (1958 Agreement) prescribe the conditions for reciprocal recognition of approvals granted on the basis of the regulations (i.e., approved once and accepted everywhere). UN Regulations are annexed to the 1958 Agreement and therefore are part of it; they are considered international law. Contracting parties are free to be bound by all, some, or no UN Regulations. UN GTRs instead do not contain administrative provisions (for certification and conformity of production). Adoption of UN GTRs needs consensus vote, while UN Regulations need a four-fifths majority of the relevant Committees.

2.1.5. The Agreements Worldwide

The countries that have signed the 1958 and 1998 Agreements can be found in the Blue Book [44]. They are referred to as contracting parties. For example, Japan and Korea have signed the 1958 Agreement, but not the USA (United States of America), Canada, China, or India. The 1998 Agreement has been signed by many countries, including the USA, Canada, Japan, Russia, China, Korea, India, and Australia.

The EU is a contracting party of the 1958 and 1998 Agreements, but not of the 1997 Agreement. In recent years, the EU decided to replace as many EU Directives as possible with the 1958 Agreement UN Regulations and to make direct reference to these UN Regulations in EU legislation [45,46]. The UN Regulations, which are applicable under EU law, must be translated into all official EU languages and published in the Official Journal of the EU.

It should be added that an EU Regulation is a binding legislative act. It must be applied in its entirety across the EU. An EU Directive is a legislative act that sets out a goal that all EU countries must achieve. However, it is up to the individual countries to devise their own laws on how to reach these goals.

2.2. Working Party on Pollution and Energy (GRPE)

The Working Party on Pollution and Energy (GRPE) is the subsidiary body of the World Forum for Harmonization of Vehicle Regulations (WP.29) that prepares regulatory proposals on pollution and energy efficiency which are submitted for approval to WP.29. Final decisions are taken by representatives of the contracting parties by vote at the World Forum WP.29 level. Some of the informal working groups (IWGs), active or inactive at the time of writing, can be found in Figure 1. Each informal working group operates on the basis of Terms of Reference (ToR) and a mandate that have to be approved by the GRPE during its plenary sessions.

The Particle Measurement Programme (PMP) group is the one responsible for the development of the technical specifications for particulate instruments and relevant regulations, and they provide feedback to other IWGs whenever requested on particulates topics. All the documents can be found on the relative website [47].

2.3. Particle Measurement Programme (PMP)

The Particle Measurement Program (PMP) group was launched in 2001 on the initiative of several UNECE contracting parties, and since then, it has evolved into an international group comprising governments, the European Commission, industry (associations of car and engine manufacturers, instrument manufacturers), and national vehicle emission laboratories and research institutions [48]. The original aim was to develop a sensitive and accurate methodology that would replace or complement the regulated gravimetric procedure, requiring low investment costs, with special consideration to particle emissions at very low levels. The PMP group was initially chaired by the UK (United Kingdom) and since 2013 by the JRC.

In the first two phases (2001–2003), a wide range of measurement techniques and sampling systems were assessed. Phase three consisted of inter-laboratory exercises (2004–2006 light-duty, 2007–2009 heavy-duty) evaluating the repeatability and reproducibility of the two final recommended systems (improved PM mass method, new particle number method). The measurement method proposed by PMP in 2007, based on the counting of solid particles with a diameter larger than 23 nm, was integrated into the European Emissions Regulation in 2011 for diesel light-duty vehicles (Euro 5b) and in 2013 for heavy-duty engines (Euro VI). The methodology was also added in UN Regulation 83 and UN Regulation 49.

PMP group activities stopped at the end of 2011 with the introduction of the method in light-duty European regulation and the proposal for heavy-duty engines. The number methodology was included in the light-duty GTR 15 (WLTP) since this was developed after the introduction of the particle number for light-duty vehicles in EU. However, heavy-duty GTR 4 (WHDC) was developed before the introduction of the particle number in the EU heavy-duty regulation, so it does not include the particle number methodology.

In 2013, the EU and Switzerland requested further investigation of particle number emissions from spark ignition engines, in particular below 23 nm, the lower size in the regulated method. The new Terms of References (ToR) included topics such as:

- Emissions from gasoline direct injection (GDI) vehicles.
- Development of equipment to measure particles below 23 nm.
- Calibration procedures update (and if necessary for lower size).
- Engine dynamometer raw exhaust particle measurements for type-approval of heavy-duty engines.
- Emissions during regeneration events.
- Non-exhaust particle emissions.

The work of the PMP group was the basis of the extension of the particle regulation to light-duty gasoline direct injection vehicles in 2014. A limit was introduced for non-road mobile machinery in 2019.

A major part of the work was the preparation of a sub-23 nm protocol. The final 10 nm proposal was introduced as an option to GTR 15 and is also ready for heavy-duty regulations. The principles that have guided the development of the sub-23 nm methodology are summarised below:

- Aim at the best compromise between accuracy, repeatability, reproducibility, and test complexity/cost.
- Try to avoid, as much as possible, requiring heavy changes and investments to update the existing facilities/measuring equipment.
- Take into account the possible different approaches in different regions (in some regions there is the intention to regulate sub-23 nm, while others could decide to do this at a later stage).

Regarding on-road particle measurements, the measurement methodology was developed in Europe by the RDE expert group and the JRC. RDE testing was introduced into EU regulation in 2017 for light-duty vehicles and in 2021 for heavy-duty vehicles. After the introduction in the EU regulation, the PMP refined and extended the methodology to 10 nm. The proposal was submitted to the UN RDE group in 2021.

An important amount of work has also been done and still continues within the PMP on defining the brake emissions protocol (cycle, sampling, instrumentation). This topic will not be addressed in this paper. The details of the particle number technical specifications for exhaust emissions will be described in the next chapter.

3. Technical Specifications

3.1. Terminology

The definition of a solid particle is not simple. The intention of the regulation is to measure “solid” particles, with the main focus being on soot particles in the past. The PMP protocol, as drafted in 2003, requires sampling from the full dilution tunnel with constant volume sampling (CVS). Thus, the exhaust gas sample is already diluted at approximately ambient temperature, around 25 °C. The decision to sample from the dilution tunnel was made because the dilution tunnels were already used for the measurement of other pollutants. For heavy-duty and non-road mobile machinery, in addition to the dilution tunnel, sampling from the proportional partial flow dilution system (PFDS) is allowed (see Figure 2). The diluted sample temperature is also expected to be low, around 25 °C. The sample preconditioning in the laboratory particle number system (LABS) includes hot dilution at >150 °C followed by an evaporation tube at 300–400 °C. Any surviving particles large enough to be detected with a particle counter having approximately 50% counting efficiency at 23 nm are defined as “solids” in the PMP protocol. These operationally defined solid particles may also include semivolatile material not evaporating at 300–400 °C (e.g., heavy molecular hydrocarbons) or at least not shrinking to a sufficiently small size that would not activate and grow to a size to be detected by the particle counter. Thus, the term “nonvolatiles” may be more appropriate; nevertheless, the commonly used term “solids” will be used in this paper.

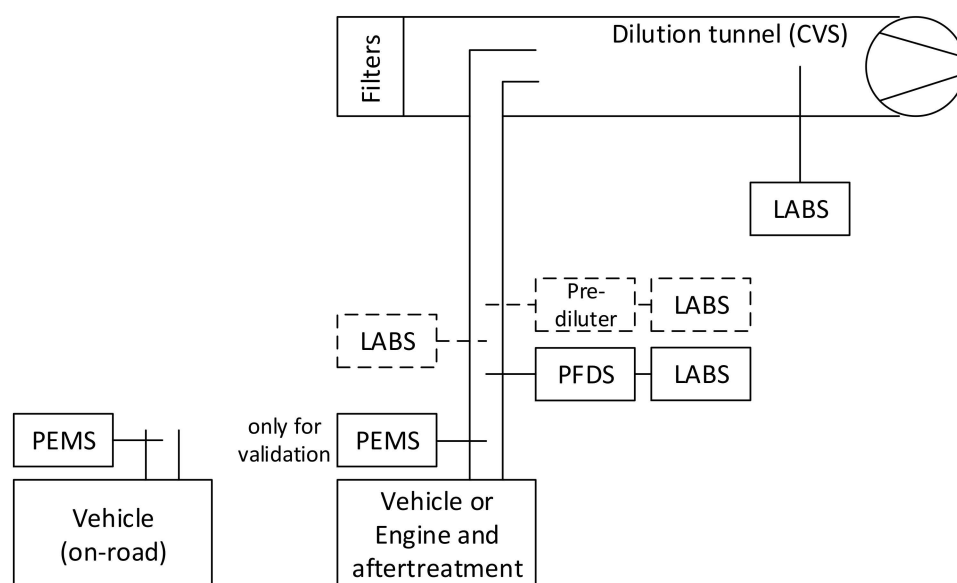


Figure 2. Sampling possibilities of laboratory particle number systems (LABS) and portable emission measurement systems (PEMS). Dashed lines show cases under discussion for inclusion in the regulation. Validation is the check of the PEMS with LABS. CVS = constant volume sampling; PFDS = proportional partial flow dilution system.

The latest terminology in the regulations (in GTR 15) uses the term “particle” for the matter being characterised (measured) in the airborne phase (suspended matter) and the term “particulate” for the deposited matter. Particle number (PN) emissions mean the total number of solid particles emitted from the vehicle exhaust quantified according to the dilution, sampling, and measurement methods as specified in the regulation. Particulate matter (PM) emissions mean the mass of any particulate material from the vehicle exhaust quantified according to the dilution, sampling, and measurement methods as specified in the regulation. UNR 49 defines as PM any material collected on a specified filter medium after diluting exhaust with a clean filtered diluent to a temperature between 42 °C and 52 °C; this is primarily carbon, condensed hydrocarbons, and sulphates with associated water. In the same regulation, instead of PN, mistakenly, the term “PM number” is used. Recently, the abbreviation SPN (solid particle number) instead of PN was suggested to emphasise that solid particles are measured and not total particles (i.e., solids and volatiles).

The on-board portable emission measurement systems (PEMS) measure directly from the tailpipe (undiluted exhaust), as the measurements are done on the road. Permitting direct sampling from the tailpipe also in the laboratory is an attractive approach. However, for light-duty vehicles, there is no exhaust gas flow rate information, which is necessary for the calculation of the emissions. On the other hand, for heavy-duty engines, direct sampling of gaseous pollutants is already permitted, and the exhaust flow rate can be measured by various methods. Thus, allowing direct particle sampling from the tailpipe for type-approval of engines is easier to introduce in the regulation. One approach is directly connecting the particle number system to the tailpipe using a heated sampling line (see Figure 2). Another is using a (cold) pre-diluter at ambient temperature, with something similar happening with CVS or the PFDS. PEMS with both concepts exist already for light-duty vehicles (i.e., cold dilution or heated line and hot dilution). Figure 2 summarises the possibilities of particle number measurements with laboratory or portable systems. Such possibilities may result in differences regarding the final SPN concentration due to incomplete removal of volatiles, particle losses (e.g., due to thermophoresis in sampling lines), and particle transformations (e.g., due to agglomeration in sampling lines). Furthermore, the optical detectors of PEMS are not necessarily the same as those

of laboratory systems, further enhancing the differences between PEMS and LABS (see Appendix A for details).

In the following sections, the term LABS will be used for the laboratory systems and PEMS for the on-board systems, followed by a number (10 or 23) indicating the approximate lower detection size (e.g., LABS₂₃ or PEMS₁₀). The term solid particle number (SPN) will be used for all the above protocols regardless of the application (laboratory or on-board). Any differences to the final SPN result due to the different design characteristics of the systems are taken into account with the limit values and the conformity factors in Regulation (EU) 2017/1151 for light-duty vehicles and Regulation (EU) 2019/1939 for heavy-duty vehicles.

3.2. Laboratory Systems (LABS) Specifications

3.2.1. Description of the System

Figure 3 plots a LABS according to the current (23 nm) technical specifications. It also gives the improvements of the 23 nm systems and the future 10 nm requirements. The laboratory systems (LABS) consist of a volatile particle remover (VPR) and a particle number counter (PNC). The VPR extracts a small flow, having a probe in a dilution tunnel (full flow or proportional partial flow). Optionally, a sampling line connects the VPR to the sampling probe. The VPR has to dilute the sample at least ten times, and the mixture temperature has to be at least 150 °C. Theoretically, a system heated at, for example, 160 °C fulfils the design requirements of the regulation (but would still have to fulfil the performance requirements for volatile removal efficiency, see below). Nevertheless, the recommended system described in the regulation, which was based on the PMP suggested system, includes an evaporation tube at 350 °C. The PNC has to be full flow and count particles in the single counting mode. Other calibration requirements regarding the particle concentration reduction factor (PCRF) and the volatile removal efficiency (VRE) of the VPR, as well as the counting efficiency (CE) of the PNC, are summarised in Figure 3. The PCRF, VRE, and CE are determined during the calibration of the device, with an appropriate aerosol measuring monodisperse in parallel with a reference instrument. For a given dilution setting of the VPR, PCRF is the ratio of the upstream to downstream concentration of solid particles of a specific size, and it is an indication of the dilution and the particle losses at the specific size for the VPR. Volatile removal efficiency is one minus the ratio of downstream to upstream concentration of volatile (tetracontane) particles with size >30 nm. Counting efficiency is the ratio of inlet to outlet concentration of particles for the PNC. Details can be found elsewhere [49].

3.2.2. Comparison of the Specifications

Table 1 summarises the technical and calibration requirements for the original laboratory system counting >23 nm (LABS₂₃) as introduced in the regulations. The next columns summarise the improved LABS₂₃ and the LABS₁₀ based on the recent PMP recommendations. Note that the terms SPN₂₃ and SPN₁₀ are used in GTR 15. The main improvements of the LABS₂₃ systems are:

- A catalytic stripper (CS) may be included in the VPR (while in the past it was not allowed).
- The PNC calibration material should be soot or PAO (polyalphaolefin) (commonly known as emery oil), while in the past there was no material definition.
- The k-factor from the linearity test has to be included in the counting efficiencies (while in the past it was not necessary).
- The differences from the reference instrument have to be within 5% (k-factor included) and not 10% as in the past.
- There are no more restrictions on internal PNC corrections (in the past, up to 10% of coincidence correction was allowed).

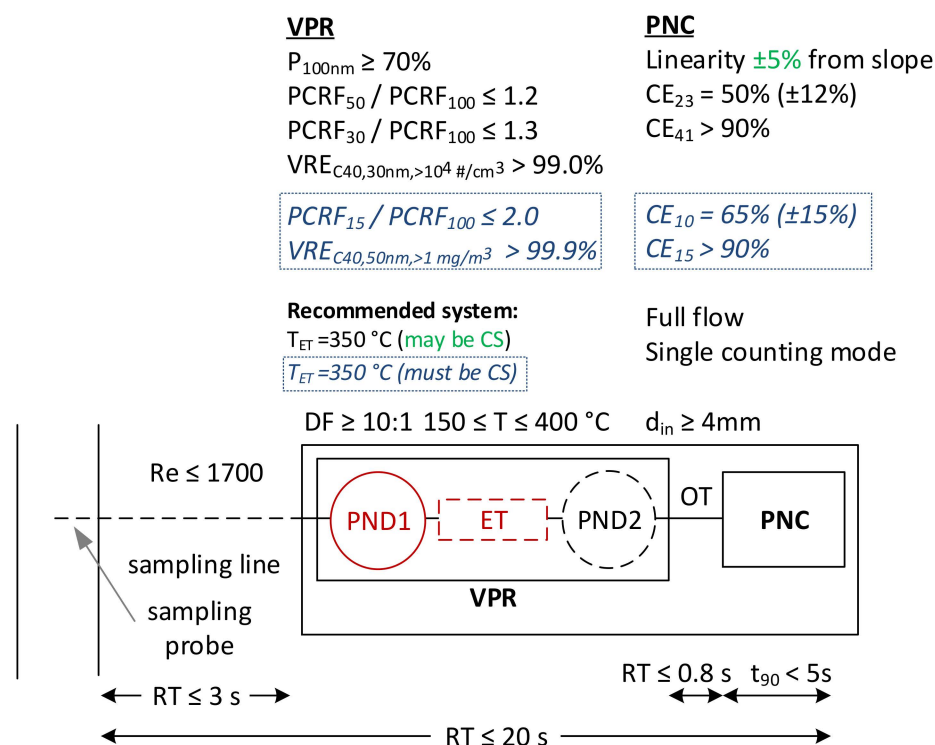


Figure 3. Schematic of a laboratory particle number system (LABS). Dashed lines show optional parts. In red are heated parts. At the top of the figure the calibration requirements are given. In green are the improvements compared to the original regulation (both counting $>23\text{ nm}$). The differences between 10-nm and 23-nm systems are given in blue italics and in dotted square. CE = counting efficiency; CS = catalytic stripper; DF = dilution factor; ET = evaporation tube; OT = outlet tube; P = penetration; PCRF = particle number concentration reduction factor; PNC = particle number counter; PND = particle number diluter; RT = residence time; t = time; T = temperature; VRE = volatile removal efficiency; VPR = volatile particle remover.

Table 1. Comparison of original LABS₂₃, improved LABS₂₃, and new (improved) LABS₁₀ technical specifications and calibration requirements. Dashed lines separate the calibration requirements.

Part	LABS ₂₃ Original	LABS ₂₃ Improved	LABS ₁₀ (Improved)
Sampling line	optional RT $\leq 3\text{ s}$ Re ≤ 1700	optional RT $\leq 3\text{ s}$ Re ≤ 1700	optional RT $\leq 3\text{ s}$ Re ≤ 1700
Volatile Particle Remover (VPR)	PND1 (DF $\geq 10:1$) 150 $\leq T \leq 400$ (± 10) $^\circ\text{C}$ 300–400 $^\circ\text{C}$ (Rec. system) No CS $T_{\text{in,PNC}} < 35^\circ\text{C}$ $P_{100} \geq 70\%$ (each instrument) $\text{PCRF}_{50}/\text{PCRF}_{100} \leq 1.2$ $\text{PCRF}_{30}/\text{PCRF}_{100} \leq 1.3$ - $\text{VRE}_{\text{C40,30nm}, \geq 10^4 \text{ \#}/\text{cm}^3} > 99.0\%$ VRE yearly Thermally stable material Calibration 12 months PCRF validation 30, 50, 100 nm or polydisperse (50 nm): $\pm 10\%$	PND1 (DF $\geq 10:1$) 150 $\leq T \leq 400$ (± 10) $^\circ\text{C}$ 350 $^\circ\text{C}$ (Rec. system) May be CS $T_{\text{in,PNC}} < \text{PNC specs}$ $P_{100} \geq 70\%$ (once for family) $\text{PCRF}_{50}/\text{PCRF}_{100} \leq 1.2$ $\text{PCRF}_{30}/\text{PCRF}_{100} \leq 1.3$ - $\text{VRE}_{\text{C40,30nm}, 10^4 \text{ \#}/\text{cm}^3} > 99.0\%$ VRE according to manuf. Thermally stable material Calibration 13 months PCRF validation 30, 50, 100 nm or polydisperse (50 nm): $\pm 10\%$	PND1 (DF $\geq 10:1$) 150 $\leq T \leq 400$ (± 10) $^\circ\text{C}$ 350 $^\circ\text{C}$ (Rec. system) With CS (Rec. system) $T_{\text{in,PNC}} < \text{PNC specs}$ $P_{100} \geq 70\%$ (once for family) $\text{PCRF}_{50}/\text{PCRF}_{100} \leq 1.2$ $\text{PCRF}_{30}/\text{PCRF}_{100} \leq 1.3$ $\text{PCRF}_{15}/\text{PCRF}_{100} \leq 2.0$ $\text{VRE}_{\text{C40}, \geq 50\text{nm}, 1 \text{ mg}/\text{m}^3} > 99.9\%$ VRE according to manuf. Thermally stable material Calibration 13 months PCRF validation 30, 50, 100 nm: $\pm 10\%$

Table 1. Cont.

Part	LABS ₂₃ Original	LABS ₂₃ Improved	LABS ₁₀ (Improved)
Particle Number Counter (PNC)	Full flow $t_{90} < 5$ s	Full flow $t_{90} < 5$ s	Full flow $t_{90} < 5$ s
	Single counting mode	Single counting mode	Single counting mode
	Any material	Soot or PAO	Soot or PAO
	$0.9 < k_{\text{slope}} < 1.1$, $R^2 > 0.97$	$0.9 < k_{\text{slope}} < 1.1$, $R^2 > 0.97$	$0.9 < k_{\text{slope}} < 1.1$, $R^2 > 0.97$
	Linearity $\pm 10\%$	Linearity $\pm 5\%$ from slope	Linearity $\pm 5\%$ from slope
	$CE_{23} = 50\%$ ($\pm 12\%$)	$CE_{23} = 50\%$ ($\pm 12\%$)	$CE_{10} = 65\%$ ($\pm 15\%$)
	$CE_{41} > 90\%$	$CE_{41} > 90\%$	$CE_{15} > 90\%$
	k_{slope} may be included in CE	k_{slope} included in CE	k_{slope} included in CE
Combined	Coincidence correction $< 10\%$	Any internal correction	Any internal correction
	Certificate 12 months	Certificate 13 months	Certificate 13 months
	PCR _F of 30, 50, 100 nm	PCR _F of 30, 50, 100 nm	PCR _F of 30, 50, 100 nm
	Total RT ≤ 20 s	Total RT ≤ 20 s	Total RT ≤ 20 s
Checks	Daily zero VPR < 0.5 #/cm ³	Daily zero VPR < 0.5 #/cm ³	Daily zero VPR < 0.5 #/cm ³
	Daily zero PNC ≤ 0.2 #/cm ³	Daily zero PNC ≤ 0.2 #/cm ³	Daily zero PNC ≤ 0.2 #/cm ³
	Daily amb. PNC: > 100 #/cm ³	No error	No error
	Monthly flow PNC $\pm 5\%$ nominal	Flow $\pm 5\%$ last certificate	Flow $\pm 5\%$ last certificate
	6 mos. monitor or wick exchange	6 mos. monitor or wick exchange	6 mos. monitor or wick exchange
	or $\pm 10\%$ of PNC _{Ref} or ≥ 2 PNCs	or $\pm 10\%$ of PNC _{Ref} or ≥ 2 PNCs	or $\pm 10\%$ of PNC _{Ref} or ≥ 2 PNCs

amb. = ambient air; CE = counting efficiency; CS = catalytic stripper; DF = dilution factor; ET = evaporation tube; mo = months; P = penetration; PCR_F = particle number concentration reduction factor; PNC = particle number counter; Rec. = recommended; RT = residence time; T = temperature; VRE = volatile removal efficiency; VPR = volatile particle remover.

The main differences between the LABS₁₀ and improved LABS₂₃ are:

- The counting efficiencies of the PNC are different and shifted to smaller particles.
- The CS is obligatory in the VPR (but only for the recommended system).
- The volatile removal efficiency has to be $> 99.9\%$ with a challenging aerosol of 1 mg/m³ tetracontane.

Details can be found in Table 1.

3.2.3. Pre-Diluter

For heavy-duty particle number sampling from the tailpipe, two approaches are allowed in the proposed Resolution:

1. Using the LABS, but the sampling line must be heated at a temperature ≥ 150 °C, and the residence time must be ≤ 1 s.
2. Using a pre-diluter and then the LABS.

Pre-diluter is defined as a diluter located after the sampling probe and in front of the sampling line of the VPR (thus, it is not part of the VPR) (Table 2). A fixed dilution ratio of > 5.1 is required, and the diluter temperature has to be ≥ 20 °C. If there is a sampling line between pre-diluter and sampling probe, it must be heated at a temperature ≥ 150 °C and have a residence time ≤ 1 s. The current proposal for the calibration requirements is that the pre-diluter and the sampling line after the VPR shall fulfil the penetration and PCR_F requirements of the LABS with an approximately 10% “worsening” of the performance.

Table 2. Technical specifications of the pre-diluter.

Specification	Description
Location	A cold or hot pre-diluter may be located at the end of the particle sampling probe and in front of the particle transfer tube (PTT). ¹
Dilution	A fixed dilution ratio >5:1 shall be applied to the cold or hot dilution stage. Cold dilution is defined as a dilution with (unheated) dilution air and/or diluter temperature ≥ 20 °C
Penetration	The complete system (pre-diluter, PTT, and VPR) penetration shall not decrease more than 10% the penetration requirements specified for the VPR.
PCRF	The complete system (pre-diluter, PTT, and VPR) PCRFs shall not exceed 0% of the PCRF requirements specified for the VPR for 50 nm, 10% for 30 nm, and 25% for 15 nm (if applicable).

¹ The residence time until the pre-diluter shall be ≤ 1 s. The tubing shall be heated at ≥ 150 °C if ≥ 10 cm

3.3. Portable Emission Measurement Systems (PEMS) Specifications

The requirements for the on-board SPN₂₃ portable emissions measurement systems (PEMS₂₃) were developed in the EU Real-Driving Emissions (RDE) group (the term “PN analyzer” is used in the regulation). The specifications had to be strict enough to be comparable with the laboratory systems but “relaxed” enough to allow small and light systems or novel concepts (e.g., hot condensation particle counters) [50] (Figure 4). In order to allow possible future innovative concepts, the main requirements for the PEMS₂₃ were to:

- Include a heated section of at least 300 °C.
- Have a volatile removal efficiency of at least 99% for 1 mg/m³ tetracontane particles with count median diameter (CMD) ≥ 50 nm.
- Be linear, within $\pm 15\%$ of a reference instrument.
- Have the efficiencies of Table 3. Efficiency is the ratio of the downstream to upstream concentration of solid particles with a specific size, as determined during the calibration of the system. The instruments have to take into account the internal dilution and any other correction factors for losses or the particle detector. These efficiencies were based on simulations of laboratory systems LABS₂₃ (combination of VPR and PNC) and an additional flexibility for on-board systems.

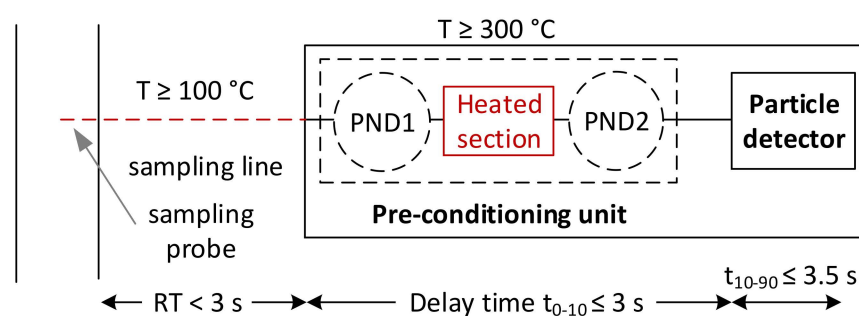


Figure 4. Schematic of a particle number portable emissions measurement system (PEMS). Dashed lines show optional parts. In red are heated parts. PND = particle number diluter; RT = residence time; t = time; T = temperature.

Table 3. Efficiencies of particle number portable emissions measurement systems (PEMS).

System	10 nm	15 nm	23 nm	30 nm	50 nm	70 nm	100 nm	200 nm
PEMS ₂₃			0.20–0.60	0.30–1.20	0.60–1.30	0.70–1.30	0.70–1.30	0.50–2.00
PEMS ₁₀	0.10–0.50	0.30–0.70		0.75–1.05	0.85–1.15	0.85–1.15	0.80–1.20	0.80–2.00

The terminology of the on-board systems is also slightly different in order to not be confused with the laboratory systems. For example, the particles are counted with a “particle detector” that could be a CPC (condensation particle counter)- or a DC (diffusion

charger)-based instrument. In contrast, in a laboratory system, the particles are counted with the PNC, which is CPC-based. A VPR, which is called “pre-conditioning system” (e.g., it could be part of the particle detector), is not obligatory in the PEMS. The sampling line, as with the laboratory systems, is not obligatory. If used, though, it has to be heated at ≥ 100 °C (while there is no temperature requirement for the laboratory systems because they measure diluted exhaust).

The SPN PEMS calibration has to be done annually with soot-like particles. For the efficiencies (Table 3), monodisperse particles have to be used, and the correction for multiply charged particles must be $<10\%$ (and reported). For the linearity, monodisperse (≥ 45 nm) or polydisperse aerosol (CMD ≥ 45 nm) can be used, and at least five concentrations covering the whole measurement range have to be checked. The slope has to be within 0.85 and 1.15 with $R^2 > 0.95$. The offset has to be $\leq 10\%$ of $x_{\max}$ and standard error of the estimate (SEE) $\leq 10\%$ of $x_{\max}$. The zero level has to be <5000 #/cm³ (averaging of 30 s) and remain <5000 #/cm³ (averaging of 30 s) after 4 h of testing. Note that the 30 s averaging will change to 60 s in order to be more in line with the ISO 27891 for CPCs.

The differences between the 10 nm to the 23 nm system are:

- The efficiencies are different (see Table 3), and the tolerances are tighter for the 10 nm systems.
- The heated part must include a catalytic stripper.
- The volatile removal efficiency has to be at least 99.9% (instead of 99.0%).

At the time of writing, the 10 nm proposal is for the light-duty UN RDE, but they will be the same for heavy-duty applications (see Discussion).

3.4. Summary of SPN Regulations

The following sections summarise the SPN regulations in the EU. Note that up until November 2009, all EU legislation references the European Community (EC), as only this body had legal personality. Since 1 December 2009, the EU is referenced, because due to the Lisbon Treaty, the European Union (EU) has been given a single legal personality.

3.4.1. Light-Duty Vehicles

In the EU, a SPN₂₃ limit of 6×10^{11} #/km applied for the first time in 2011 (Euro 5b) for diesel vehicles (Regulation (EC) 692/2008) (Table 4). Regulation (EU) 459/2012 introduced the same limit for GDIs in 2014 (Euro 6), with the possibility of a 10-times-higher limit for the first 3 years, upon request of the vehicle manufacturer. Regulation (EU) 2017/1151 (Euro 6c) repealed Regulation (EC) 692/2008 and introduced the worldwide harmonised light vehicles test procedure, or WLTP. Real-driving emissions (RDE) testing with portable emissions measurement systems (PEMS) was introduced in 2017 (Euro 6d-Temp) initially only for type-approval (Regulations (EU) 2016/427, 2016/646), with a conformity factor (CF) of 1.5 for particle number (Regulation (EU) 2017/1154), and later for in-service conformity (ISC) (Regulation (EU) 2018/1832). The CF for on-road SPN₂₃ with PEMS₂₃ remained the same for Euro 6d, which entered into force in 2020. All RDE regulations were consolidated in Regulation (EU) 2017/1151. 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).

Table 4. Regulations for light-duty vehicles regarding type-approval, conformity of production, and in-service conformity. The limits are applicable to the relevant EU and UNR regulations. GTRs do not include any emission limits.

Location	GTR	UNR	EU	SPN ₂₃ (#/km)	PM (mg/km)
LABS ₂₃	15	83 or 154	2017/1151	6×10^{11}	4.5
PEMS ₂₃	- ¹	- ²	2017/1154	CF = 1.5 ³	-

¹ informal document [51]; ² working document [52]; ³ to be reduced to 1.34 [53].

3.4.2. Heavy-Duty Vehicles

In EU SPN₂₃, limits were initially applied to compression ignition (CI) (diesel) engines in 2013 (Euro VI) (Regulation (EU) 582/2011) and in 2014 to positive ignition (PI) engines (Regulation (EU) 133/2014) (Table 5). The limit is 6×10^{11} #/kWh for the combined (14% cold, 86% hot) WHTC and 8×10^{11} #/kWh for the WHSC (only diesel). The methodology (Regulation (EU) 64/2012) refers to UNR 49.

Regulation (EU) 582/2011 introduced on-road PEMS testing for ISC (instead of removing the engine from a vehicle and testing it on the engine dynamometer) for gaseous pollutants. Regulation (EU) 2019/1939 (Euro VI step E) added the cold start in the evaluation of the emissions and SPN₂₃ with a CF = 1.63 with PEMS₂₃ from 2021 for CI engines and 2023 for PI engines.

Table 5. Regulations for heavy-duty engines and vehicles regarding type-approval and in-service conformity. The limits are applicable to the relevant EU and UNR regulations. GTRs do not include any emission limits.

Location	GTR	UNR	EU	SPN ₂₃ (#/kWh)	PM (mg/kWh)
LABS ₂₃	4 (no PN)	49	582/2011	6×10^{11} ¹	10
PEMS ₂₃	-	-	582/2011	CF = 1.63	-

¹ 8×10^{11} #/kWh for the steady cycle (only diesel).

3.4.3. Non-Road Mobile Machinery

In the EU, Regulation (EU) 2016/1628 repealed Directive 97/68/EC in 2016 and introduced SPN₂₃ limits to non-road engines (19–560 kW), inland waterway vessels (>300 kW), and rail traction engines in 2019 (Stage V) (Table 6). The procedures and test cycles (non-road transient cycle—NRTC; non-road stationary cycle—NRSC) are described in Regulation (EU) 2017/654 and 2017/655 for on-road monitoring. At the moment, there is no particle number limit for on-road testing with PEMS.

Table 6. Regulations for non-road mobile machinery engines and vehicles regarding type-approval and in-service conformity. The limits are applicable to the relevant EU and UNR regulations. GTRs do not include any emission limits.

Location	GTR	UNR	EU	SPN ₂₃ (#/kWh)	PM (mg/kWh)
LABS ₂₃	11 (no PN)	96	2017/654	1×10^{12}	15
PEMS ₂₃	-	-	2017/655 ¹	-	-

¹ no SPN for on-road testing.

3.4.4. Worldwide

Worldwide, many countries in Asia have introduced SPN₂₃ limits in the laboratory (6×10^{11} #/km for light-duty vehicles, 6×10^{11} #/kWh for heavy-duty engines) in the last few years (Table 7). On-road testing will be required from 2023 in India (CF to be defined) and China (CF = 2.0–2.1). Korea has limits only for diesel vehicles [54] whereas India also includes gasoline vehicles with direct injection engines, and China limits all technologies (with CN 6).

Table 7. Summary of regulations including particle number limits. The table gives the first emission stage including a particle number limit. In brackets, the relative technology and the introduction year.

Country	System	Light-Duty	Heavy-Duty	NRMM
Korea	LABS ₂₃	Euro 6 (D, 2014)	Euro VI (D, 2015)	no
Singapore	LABS ₂₃	Euro 6 (D, GDI 2018)	Euro VI (all, 2018)	no
India	LABS ₂₃	BS VI (D, GDI, 2020)	BS VI (all, 2020) ¹	BS V (2024)
	PEMS ₂₃	CF = tbd (2023)	CF = tbd (2023)	
China	LABS ₂₃	CN5 (D, 2016) ²		
	LABS ₂₃	CN 6a (all, 2020)	CN VIa (all, 2019)	Stage IV (2023) ¹
	PEMS ₂₃	CN 6b (CF = 2.1, 2023)	CN VIb (CF = 2.0, 2021)	CF = 2.5 (no SPN)

¹ 2025 for positive ignition; ² earlier in Beijing (2013) and Shanghai (2014); limit 5×10^{12} p/kWh. BS = Bharat stage; CN = China; D = diesel; GDI = gasoline direct injection; NRMM = non-road mobile machinery.

4. Discussion

4.1. Open Points

The open topics of the PMP group regarding exhaust emissions part are described below.

One is the investigation of whether the volatile removal efficiency test is adequate. The tetracontane is representative of heavy molecular hydrocarbons, and 1 mg/m^3 is a mass at the high end of what is typically seen at the exhaust of modern engines [55]. Higher values have been measured for two-stroke mopeds or vehicles without oxidation catalyst. Hydrocarbons are rarely the nucleating species, e.g., with engines without oxidation catalysts [56] where the concentrations are high. For engines with oxidation catalysts, sulfuric acid is the key compound in forming the first nuclei [57]. The nucleation can be enhanced when a small amount of ammonia is present [58]. Hydrocarbons are necessary, though, to grow the particles to the $>10 \text{ nm}$ range. Thus, the current volatile removal requirement ensures that the measurement systems are capable of reducing the nucleation mode to sizes not detectable by the particle detectors. Another difficulty is the requirement of a generated mean size of $\geq 50 \text{ nm}$, which has been found to result in residual particles. Discussions are ongoing to reduce this requirement to 30 nm .

Another important topic is the improvement of the calibration procedures. More detailed uncertainty analysis and traceability have to be re-examined. Even though a lot of work has been done in the last few years [33,59–63], the different specifications between laboratory and on-board systems make the comparison of the results of the two systems difficult. Any estimation of the additional uncertainty of PEMS vs. LABS needs to estimate the efficiencies of the LABS based on the VPR particle concentration reduction factors and PNC counting efficiencies [53]. For example, defining common calibration sizes would improve the comparability.

Another point that needs more research is the pre-diluter. Most studies have been done with direct sampling and a heated line and showed acceptable agreement with the current PFDS or CVS systems [64,65]. The results with the pre-diluter tend to slightly overestimate the emissions. This needs to be better understood. Furthermore, the tests so far have had exhaust gas temperatures $<500 \text{ }^\circ\text{C}$, and it is not known how the systems will behave at higher temperatures.

4.2. Future SPN Regulations

The improved LABS₂₃ and LABS₁₀ methodologies have been included in the latest amendments to the GTR 15 (light-duty). There is also a Consolidate Resolution ready for heavy-duty regulations which, in addition to the improved LABS₂₃ and LABS₁₀ methodologies, expands the LABS for sampling directly from the tailpipe. The different approach in terms of legislative acts at the UNECE level (GTR vs. Consolidated Resolution) for HD engines is linked to the fact that the particle number method for HD is described only in UNR 49 and not in GTR 4. Therefore, any change to UNR 49 would have an immediate regulatory effect. Nevertheless, the plan is to include both LABS₂₃ and LABS₁₀ options

and direct tailpipe sampling in a future amendment of GTR 4. Inclusion of improved LABS₂₃ and LABS₁₀ in UNR 49 (and UNR 83 and 154 for light-duty vehicles) still needs to be discussed. Furthermore, direct sampling from the tailpipe with fixed dilution was prepared for the type-approval of heavy-duty engines. The plan is to include this option in both GTR 4 and UNR 49.

The PEMS₁₀ methodology will be included in the future in the GTR RDE light-duty regulation. It is expected to have the same requirements for heavy-duty testing, but at the moment, there is no GTR or UNR foreseen for on-road heavy-duty vehicles testing. There is no official announcement regarding whether other countries will introduce the 10 nm methodologies (laboratory or on-road) in their regulations.

Other future particle number regulations are discussed in the next paragraphs.

4.2.1. Euro 7/VII

Since 2018, the Commission has started the development of a future-proof emissions regulation, i.e., the Euro 7. During the process of consultation, various organisations have commented on some aspects of the current EU regulation and/or had suggestions for the future regulations [66–68]. The main ones are discussed below.

The 23 nm lower size does not always cover the whole or the largest part of the size distribution of the emitted particles. High amounts of sub-23 nm particles are measured very often, both for diesel [64] and gasoline direct injection vehicles [31], but in particular for the technologies not subject currently to a SPN limit (port fuel injection gasoline and gas engines) [69–71]. On the other hand, for heavy-duty vehicles, all technologies are subject to the same limit. For this reason, it has been suggested to both (i) lower the 23 nm to 10 nm and (ii) include all technologies in the limits without exceptions.

The current testing procedures for light-duty vehicles only partly include emissions during the regeneration of periodically regenerating systems. For gaseous pollutants, the emissions during regeneration are taken into account based on the regeneration frequency. However, SPN emissions during regeneration events and immediately after are very high and can bring the weighted emissions close to the current limit. Thus, filtration efficiencies and regeneration strategies need to be carefully assessed for future regulation [72–77].

For heavy-duty vehicles, there are a few more challenges. For example, the emissions from the crankcase ventilation (when it's an open system) currently have to be included at the tailpipe emissions. However, the emission levels only from the crankcase ventilation are one-third of the current limit, and addressing them is very important in case of limit reduction [64,65]. One should also consider that urea injection can more than double the concentration of particles >23 nm and increase the sub-23 nm fraction if not properly controlled [78,79].

Finally, combining challenging situations in one trip (still within the boundaries of the regulation), e.g., cold start, low ambient temperatures [80], use of auxiliaries [81], dynamic driving [80,82], and regeneration [83], could result in very high emissions (per distance), and future vehicles will have to be appropriately calibrated.

4.2.2. L-Category

Regarding SPN limits for L-category vehicles, there has been no discussion of SPN limits since the studies for the introduction of Euro 4 and Euro 5 emission limits [84]. Studies for L-category vehicles showed that measuring only >23 nm misses a large part of the emitted particles [85]. On the other hand, due to high exhaust gas temperatures, measurements of particles >10 nm were subject to artefacts [86]. Recent studies showed that this can be avoided with the appropriate system and sampling configuration [87,88]. It is important to assess the emission levels of the recently introduced Euro 5 vehicles compared to the future Euro 7 light-duty vehicles [89]. On one hand, it is expected that the SPN levels will drop due to the stringent Euro 5 levels for gaseous pollutants. On the other hand, experience showed that not imposing any limit in the regulation does not necessarily

improve the emission levels. For example, the SPN levels of gasoline vehicles with port fuel injection engines have remained the same over the last 3–4 decades [29].

4.2.3. Periodic Technical Inspection

The previous discussion focused on methodology and instrumentation for type-approval, in-service conformity (in-use compliance), and market surveillance purposes, which fall under the responsibility of the OEMs (original equipment manufacturers) and the respective authorities. This paragraph discusses the periodic inspection of vehicles under the responsibility of each car owner. Traditionally, this test measured the exhaust opacity during unloaded acceleration from idle to a high engine speed [26]. Opacity is not sensitive enough for DPF (diesel particulate filter)-equipped vehicles, and other methods have been investigated over the years. The better sensitivity of DCs and CPCs led to the introduction of simplified SPN methodologies for the vehicles' inspection [90].

The first to introduce the particle number in inspection was Switzerland for construction machinery. For light-duty and heavy-duty vehicles, the Netherlands and Belgium will also introduce a simplified SPN methodology in July 2022, and Germany in January 2023. The methodology prescribes a measurement at idle with a sensor that detects solid particles >23 nm. The technical specifications of the instruments were based on the technical specifications of the instruments for construction machinery in Switzerland and the on-board systems of the European real-driving emissions (RDE) regulation (i.e., PEMS). The work was conducted by the informal new periodic technical inspection (NPTI) technical working group consisting of Swiss, German, and Dutch governmental organisations, the VERT (Verification of Emission Reduction Technologies) association, metrological institutes, scientists, and equipment manufacturers. The JRC of the European Commission is also working on the topic to prepare common recommendations for particle number instrumentation and procedures for PTI purposes. Details about the simplified methodology and the specifications can be found elsewhere [91].

4.2.4. Brake Emissions

Due to the low exhaust emissions from modern vehicles, the contribution of traffic-related non-exhaust emissions to air pollution has become relatively more important and is likely to increase [92,93]. In June 2021, GRPE mandated the PMP group to develop a GTR on brake emissions from light-duty vehicles. The draft is expected to be ready in June 2022, with most probable adoption in January 2023 at GRPE. The sampling methodology and the instruments' technical specifications are already under development. In addition to mass, a particle number limit of total particles (i.e., including volatiles) >10 nm is expected. The topic is out of the scope of this paper.

5. Conclusions

The PMP (Particle Measurement Programme) working group of the UNECE (United Nations Economic Commission for Europe) GRPE (Working Party on Pollution and Energy) developed the solid particle number (SPN) methodology that was introduced in various regulations. The methodology includes a heated section and counting of particles with diameter >23 nm (SPN_{23}). A SPN_{23} limit was initially introduced in the European laboratory type-approval regulation for light-duty diesel vehicles in 2011 (6×10^{11} #/km). The methodology was expanded to light-duty gasoline direct injection vehicles (2014), heavy-duty engines (6×10^{11} #/kWh) (2013), and non-road mobile machinery (1×10^{12} #/kWh) (2019). A test for on-road light-duty SPN_{23} emissions was introduced in 2017 and for heavy-duty vehicles in 2021.

UN regulations that include the particle number methodology with emission limits are UN Regulation 83 and UN Regulation 154 for light-duty vehicles and UN Regulation 49 for heavy-duty engines. The only Global Technical Regulation that includes the particle number methodology (without emission limits) is GTR 15 for light-duty vehicles. GTR 4 for heavy-duty vehicles and GTR 11 for non-road mobile machinery will probably follow. UN

regulations for on-road testing are under development and will also include the particle number methodology.

The PMP group extended the methodology to cover particles larger than 10 nm (SPN₁₀). The SPN₁₀ option (i.e., in addition to the SPN₂₃ method) is at the moment included only in GTR 15 for laboratory testing of light-duty vehicles. A heavy-duty resolution was also prepared including the 10 nm methodology and the option to measure directly from the tailpipe. The UN Regulation for on-road testing will also include it in the future. The major differences between the 10 nm and 23 nm methodologies, other than the different lower detection sizes, are the obligatory inclusion of a catalytic stripper and the stricter volatile removal efficiency requirements. Improvements to both methodologies focused on the particle number counter, namely the definition of the calibration material, inclusion of the calibration factor in the counting efficiencies, and differences from the calibration reference instrument within $\pm 5\%$ (instead of $\pm 10\%$). The PMP work is moving forward with final refinements of the calibration procedures and possibly bringing them closer for laboratory and on-board systems.

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Abbreviations

BS	Bharat stage
CE	counting efficiency
CF	conformity factor
CI	compression ignition
CMD	count median diameter
CN	China
COVID	coronavirus disease
CPC	condensation particle counter
CS	catalytic stripper
CVS	constant volume sampling
D	Diesel
DC	diffusion charger
DF	dilution factor
DPF	diesel particulate filter
EC	European Community
ECE	Economic Commission for Europe
EPPR	environmental and propulsion performance of L-category vehicles
ET	evaporation tube
EU	European Union
EVE	electric vehicles and environment
GDI	gasoline direct injection
GR	Group of Rapporteurs

GRB	Working Party on Brakes
GRBP	Working Party on Noise and Tyres
GRE	Working Party on Lighting and Light-Signalling
GRP	Working Party on Passive Safety
GRPE	Working Party on Pollution and Energy
GRRF	Working Party on Brakes and Running Gear
GRS	Working Party on General Safety Provisions
GRV	Working Party on Automated/Autonomous and Connected Vehicles
GTR	Global Technical Regulation
ISC	in-service conformity
IWG	informal working group
NGO	non-governmental organisations
NPTI	new periodic technical inspection
NRMM	non-road mobile machinery
NRSC	non-road stationary cycle
NRTC	non-road transient cycle
OEM	original equipment manufacturer
OT	outlet tube
P	penetration
PAO	polyalphaolefin
PCRF	particle number concentration reduction factor
PEMS	portable emissions measurement system
PFDS	proportional partial flow dilution system
PI	positive ignition
PM	particulate matter
PMP	Particle Measurement Programme
PN	particle number
PNC	particle number counter
PND	particle number diluter
PTI	periodic technical inspection
RDE	real-driving emissions
Re	Reynolds number
REC	retrofit emissions control devices
RT	residence time
SEE	standard error of the estimate
SPN	solid particle number
T	temperature
t	time
ToR	terms of reference
UN	United Nations
UNECE	United Nations Economic Commission for Europe
UNR	United Nations regulation
USA	United States of America
VERT	verification of emission reduction technologies
VIAQ	vehicle interior air quality
VPR	volatile particle remover
VRE	volatile removal efficiency
WHDC	worldwide harmonised heavy-duty certification procedure
WHO	World Health Organization
WHSC	worldwide harmonised stationary cycle
WHTC	worldwide harmonised transient cycle
WLTC	worldwide harmonised light vehicles test cycle
WLTP	worldwide harmonised light vehicles test procedure
WMTC	worldwide harmonised motorcycle test cycle
WP.29	World Forum for Harmonization of Vehicle Regulations

Appendix A

Figure A1 plots the principle of operation of the systems for the measurement of solid particles. The upper part shows a volatile particle remover (VPR) with an evaporation tube, followed by a particle number counter (PN) based on a condensation particle counter (CPC) [59]. The lower part shows a VPR with a catalytic stripper [55], followed by a diffusion charger (DC) based particle detector [94,95]. The schematic shows how particles evolve in the different parts.

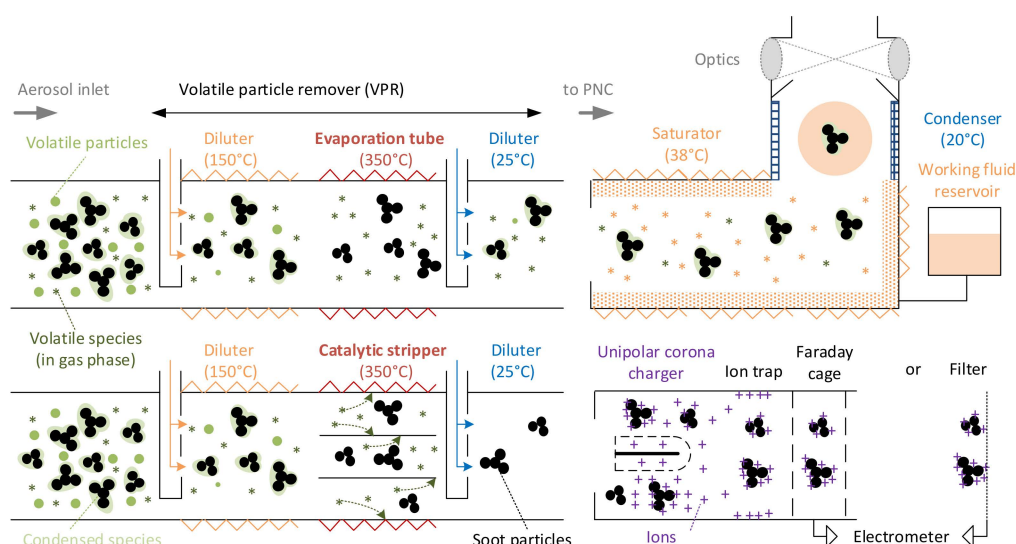


Figure A1. Schematic presentation of the principles of measurements of solid particles. The upper part shows a volatile particle remover (VPR) with evaporation tube, followed by a particle number counter (PNC) based on condensation particle counter (CPC). The lower part shows a VPR with catalytic stripper, followed by a diffusion charger with a Faraday cage and pulsed current modulation or a final filter. Schematic based on [26,55,95].

Starting with the upper left part of the figure, the aerosol at the inlet of the VPR consists of soot particles with some condensed material on them, volatile nucleation mode particles (green circles), and volatile species in the gaseous phase (asterisks) [26]. In the hot diluter of the VPR, the concentration of particles is reduced and some volatiles evaporate. Complete evaporation takes place in the evaporation tube. During the secondary cold dilution, some of the volatiles condense on the soot particles. The rest have low partial pressures and cannot nucleate to form volatile particles. In the CPC, the soot particles pass from the saturator, where butanol (or propanol) vapours exist. Then, in the condenser part, due to the rapid cooling, the vapours condense on the particles and enlarge them from the nanometre range to the micrometre range to be counted by the laser optics of the CPC.

The lower part of Figure A1 plots another case with a catalytic stripper. The catalytic stripper oxidises the volatile species, so after the secondary diluter, only the soot particles remain. After the VPR, a PNC, as previously described, could be connected. In this example, there is a diffusion charger-based particle detector. The particles pass through a corona charger and the produced ions attach on the particles. An ion trap removes the rest of the ions. The charged particles deposit on a filter or are captured in a Faraday cage where an electrometer measures the current of the particles. The current is proportional to the active surface area of the particles, but it can also be used as an indication of the particle number concentration. Details can be found elsewhere about other concepts [94].

References

1. Fowler, D.; Brimblecombe, P.; Burrows, J.; Heal, M.R.; Grennfelt, P.; Stevenson, D.S.; Jowett, A.; Nemitz, E.; Coyle, M.; Liu, X.; et al. A Chronology of Global Air Quality. *Phil. Trans. R. Soc. A* **2020**, *378*, 20190314. [[CrossRef](#)]
2. European Commission. *Statistical Office of the European Union. Urban Europe: Statistics on Cities, Towns and Suburbs: 2016 Edition*; Publications Office: Luxembourg, 2016.
3. Molina, L.T. Introductory Lecture: Air Quality in Megacities. *Faraday Discuss.* **2021**, *226*, 9–52. [[CrossRef](#)]
4. Health Effects Institute. *State of Global Air 2020*; Health Effects Institute: Boston, MA, USA, 2020.
5. Pope, C.A.; Coleman, N.; Pond, Z.A.; Burnett, R.T. Fine Particulate Air Pollution and Human Mortality: 25+ Years of Cohort Studies. *Environ. Res.* **2020**, *183*, 108924. [[CrossRef](#)]
6. Gentner, D.R.; Jathar, S.H.; Gordon, T.D.; Bahreini, R.; Day, D.A.; El Haddad, I.; Hayes, P.L.; Pieber, S.M.; Platt, S.M.; de Gouw, J.; et al. Review of Urban Secondary Organic Aerosol Formation from Gasoline and Diesel Motor Vehicle Emissions. *Environ. Sci. Technol.* **2017**, *51*, 1074–1093. [[CrossRef](#)]
7. Simonen, P.; Kalliokoski, J.; Karjalainen, P.; Rönkkö, T.; Timonen, H.; Saarikoski, S.; Aurela, M.; Bloss, M.; Triantafyllopoulos, G.; Kontses, A.; et al. Characterization of Laboratory and Real Driving Emissions of Individual Euro 6 Light-Duty Vehicles—Fresh Particles and Secondary Aerosol Formation. *Environ. Pollut.* **2019**, *255*, 113175. [[CrossRef](#)]
8. Shaddick, G.; Thomas, M.L.; Mudu, P.; Ruggeri, G.; Gummy, S. Half the World’s Population Are Exposed to Increasing Air Pollution. *NPJ Clim. Atmos. Sci.* **2020**, *3*, 23. [[CrossRef](#)]
9. Belis, C.A.; Karagulian, F.; Larsen, B.R.; Hopke, P.K. Critical Review and Meta-Analysis of Ambient Particulate Matter Source Apportionment Using Receptor Models in Europe. *Atmos. Environ.* **2013**, *69*, 94–108. [[CrossRef](#)]
10. Lorelei de Jesus, A.; Thompson, H.; Knibbs, L.D.; Kowalski, M.; Cyrus, J.; Niemi, J.V.; Kousa, A.; Timonen, H.; Luoma, K.; Petäjä, T.; et al. Long-Term Trends in PM_{2.5} Mass and Particle Number Concentrations in Urban Air: The Impacts of Mitigation Measures and Extreme Events Due to Changing Climates. *Environ. Pollut.* **2020**, *263*, 114500. [[CrossRef](#)] [[PubMed](#)]
11. European Environment Agency. *Air Quality in Europe: 2020 Report*; Publications Office: Luxembourg, 2020.
12. Donzelli, G.; Cioni, L.; Cancellieri, M.; Llopis Morales, A.; Morales Suárez-Varela, M. The Effect of the COVID-19 Lockdown on Air Quality in Three Italian Medium-Sized Cities. *Atmosphere* **2020**, *11*, 1118. [[CrossRef](#)]
13. Fu, F.; Purvis-Roberts, K.L.; Williams, B. Impact of the COVID-19 Pandemic Lockdown on Air Pollution in 20 Major Cities around the World. *Atmosphere* **2020**, *11*, 1189. [[CrossRef](#)]
14. Grivas, G.; Athanasopoulou, E.; Kakouri, A.; Bailey, J.; Liakakou, E.; Stavroulas, I.; Kalkavouras, P.; Bougiatioti, A.; Kaskaoutis, D.; Ramonet, M.; et al. Integrating in Situ Measurements and City Scale Modelling to Assess the COVID-19 Lockdown Effects on Emissions and Air Quality in Athens, Greece. *Atmosphere* **2020**, *11*, 1174. [[CrossRef](#)]
15. Lotrecchiano, N.; Trucillo, P.; Barletta, D.; Poletto, M.; Sofia, D. Air Pollution Analysis during the Lockdown on the City of Milan. *Processes* **2021**, *9*, 1692. [[CrossRef](#)]
16. Harrison, R.M.; Vu, T.V.; Jafar, H.; Shi, Z. More Mileage in Reducing Urban Air Pollution from Road Traffic. *Environ. Int.* **2021**, *149*, 106329. [[CrossRef](#)]
17. Pant, P.; Lal, R.M.; Guttikunda, S.K.; Russell, A.G.; Nagpure, A.S.; Ramaswami, A.; Peltier, R.E. Monitoring Particulate Matter in India: Recent Trends and Future Outlook. *Air Qual. Atmos. Health* **2019**, *12*, 45–58. [[CrossRef](#)]
18. Xiao, K.; Qin, A.; Wang, W.; Lu, S.; Wang, Q. Study on the Characteristics of Size-Segregated Particulate Water-Soluble Inorganic Ions and Potentially Toxic Metals during Wintertime in a High Population Residential Area in Beijing, China. *Processes* **2021**, *9*, 552. [[CrossRef](#)]
19. De Jesus, A.L.; Rahman, M.M.; Mazaheri, M.; Thompson, H.; Knibbs, L.D.; Jeong, C.; Evans, G.; Nei, W.; Ding, A.; Qiao, L.; et al. Ultrafine Particles and PM_{2.5} in the Air of Cities around the World: Are They Representative of Each Other? *Environ. Int.* **2019**, *129*, 118–135. [[CrossRef](#)] [[PubMed](#)]
20. Chatain, M.; Alvarez, R.; Ustache, A.; Rivière, E.; Favez, O.; Pallares, C. Simultaneous Roadside and Urban Background Measurements of Submicron Aerosol Number Concentration and Size Distribution (in the Range 20–800 nm), along with Chemical Composition in Strasbourg, France. *Atmosphere* **2021**, *12*, 71. [[CrossRef](#)]
21. Paasonen, P.; Kupiainen, K.; Klimont, Z.; Visschedijk, A.; Denier van der Gon, H.A.C.; Amann, M. Continental Anthropogenic Primary Particle Number Emissions. *Atmos. Chem. Phys.* **2016**, *16*, 6823–6840. [[CrossRef](#)]
22. Kumar, P.; Morawska, L.; Birmili, W.; Paasonen, P.; Hu, M.; Kulmala, M.; Harrison, R.M.; Norford, L.; Britter, R. Ultrafine Particles in Cities. *Environ. Int.* **2014**, *66*, 1–10. [[CrossRef](#)]
23. Rivas, I.; Beddows, D.C.S.; Amato, F.; Green, D.C.; Järvi, L.; Hueglin, C.; Reche, C.; Timonen, H.; Fuller, G.W.; Niemi, J.V.; et al. Source Apportionment of Particle Number Size Distribution in Urban Background and Traffic Stations in Four European Cities. *Environ. Int.* **2020**, *135*, 105345. [[CrossRef](#)]
24. World Health Organization. *WHO Global Air Quality Guidelines: Particulate Matter (PM_{2.5} and PM₁₀), Ozone, Nitrogen Dioxide, Sulfur Dioxide and Carbon Monoxide*; World Health Organization: Geneva, Switzerland, 2021; ISBN 978-92-4-003422-8.
25. Jeong, C.-H.; Evans, G.J.; Healy, R.M.; Jadidian, P.; Wentzell, J.; Liggio, J.; Brook, J.R. Rapid Physical and Chemical Transformation of Traffic-Related Atmospheric Particles near a Highway. *Atmos. Pollut. Res.* **2015**, *6*, 662–672. [[CrossRef](#)]
26. Giechaskiel, B.; Maricq, M.; Ntziachristos, L.; Dardiotis, C.; Wang, X.; Axmann, H.; Bergmann, A.; Schindler, W. Review of Motor Vehicle Particulate Emissions Sampling and Measurement: From Smoke and Filter Mass to Particle Number. *J. Aerosol Sci.* **2014**, *67*, 48–86. [[CrossRef](#)]

27. Giechaskiel, B.; Mamakos, A.; Andersson, J.; Dilara, P.; Martini, G.; Schindler, W.; Bergmann, A. Measurement of Automotive Nonvolatile Particle Number Emissions within the European Legislative Framework: A Review. *Aerosol Sci. Technol.* **2012**, *46*, 719–749. [CrossRef]
28. EPA. *Integrated Science Assessment for Particulate Matter*; Environmental Protection Agency: Research Triangle, NC, USA, 2019.
29. Giechaskiel, B.; Joshi, A.; Ntziachristos, L.; Dilara, P. European Regulatory Framework and Particulate Matter Emissions of Gasoline Light-Duty Vehicles: A Review. *Catalysts* **2019**, *9*, 586. [CrossRef]
30. Karjalainen, P.; Rönkkö, T.; Simonen, P.; Ntziachristos, L.; Juuti, P.; Timonen, H.; Teinilä, K.; Saarikoski, S.; Saveljeff, H.; Lauren, M.; et al. Strategies to Diminish the Emissions of Particles and Secondary Aerosol Formation from Diesel Engines. *Environ. Sci. Technol.* **2019**, *53*, 10408–10416. [CrossRef]
31. Samaras, Z.C.; Andersson, J.; Bergmann, A.; Hausberger, S.; Toumasatos, Z.; Keskinen, J.; Haisch, C.; Kontses, A.; Ntziachristos, L.D.; Landl, L.; et al. Measuring Automotive Exhaust Particles down to 10 nm. In *SAE Technical Papers*; 2020-01-2209; SAE International: Warrendale, PA, USA, 2020. [CrossRef]
32. Bischof, O.F. Recent Developments in the Measurement of Low Particulate Emissions from Mobile Sources: A Review of Particle Number Legislations. *Emiss. Control Sci. Technol.* **2015**, *1*, 203–212. [CrossRef]
33. Giechaskiel, B.; Lähde, T.; Suarez-Bertoa, R.; Clairotte, M.; Grigoratos, T.; Zardini, A.; Perujo, A.; Martini, G. Particle Number Measurements in the European Legislation and Future JRC Activities. *Combust. Engines* **2018**, *174*, 3–16. [CrossRef]
34. UNECE. *World Forum for Harmonization of Vehicle Regulations (WP.29)*; UNECE: Geneva, Switzerland, 1952; Available online: <https://unece.org/transport/vehicle-regulations/wp29-world-forum-harmonization-vehicle-regulations-wp29> (accessed on 7 December 2021).
35. UNECE. United Nations Economic Commission for Europe (UNECE). Available online: <https://unece.org/> (accessed on 7 December 2021).
36. GRPE. *Working Party on Pollution and Energy—Introduction*; UNECE: Geneva, Switzerland, 1952; Available online: <https://unece.org/transportvehicle-regulations/working-party-pollution-and-energy-introduction> (accessed on 7 December 2021).
37. UNECE. *World Forum for Harmonization of Vehicle Regulations (WP.29): How It Works, How to Join It*, 4th ed.; United Nations, Ed.; United Nations, Economic Commission for Europe: Geneva, Switzerland, 2019; ISBN 978-92-1-117189-1.
38. UNECE. *Agreement 1958 Text of the 1958 Agreement*; UNECE: Geneva, Switzerland, 1958; Available online: <https://unece.org/text-1958-agreement> (accessed on 7 December 2021).
39. UNECE. *Agreement 1998 Text of the 1998 Agreement*; UNECE: Geneva, Switzerland, 1998; Available online: <https://unece.org/text-1998-agreement> (accessed on 7 December 2021).
40. UNECE. *Agreement 1997 Text of the 1997 Agreement*; UNECE: Geneva, Switzerland, 1997; Available online: <https://unece.org/transportvehicle-regulations/text-1997-agreement> (accessed on 7 December 2021).
41. UNR 154. *UN Regulation No. 154—Worldwide Harmonized Light Vehicles Test Procedure (WLTP)*; UNECE: Geneva, Switzerland, 2021; Available online: <https://unece.org/transport/documents/2021/02/standards/un-regulation-no-154-worldwide-harmonized-light-vehicles-test> (accessed on 7 December 2021).
42. UNR. *Addenda to the 1958 Agreement (Regulations 41–60)*; UNECE: Geneva, Switzerland, 1958; Available online: <https://unece.org/un-regulations-addenda-1958-agreement> (accessed on 7 December 2021).
43. UNECE. *GTR Global Technical Regulations*; UNECE: Geneva, Switzerland, 1998; Available online: <https://unece.org/transport/standards/transport/vehicle-regulations-wp29/global-technical-regulations-gtrs> (accessed on 7 December 2021).
44. WP29 Blue Book. Available online: <https://unece.org/transport/publications/world-forum-harmonization-vehicle-regulations-wp29-how-it-works-how-join-1> (accessed on 7 December 2021).
45. European Union. International Technical Harmonisation. Available online: https://ec.europa.eu/growth/sectors/automotive/technical-harmonisation/international_en (accessed on 7 December 2021).
46. European Commission, Enterprise and Industry Directorate-General. *Cars 21: A Competitive Automotive Regulatory System for the 21st Century; Final Report*; Enterprise and Industry; Office for Official Publications of the European Communities: Luxembourg, 2006; ISBN 978-92-79-00762-0.
47. PMP Particle Measurement Programme. Available online: <https://wiki.unece.org/pages/viewpage.action?pageId=2523173> (accessed on 7 December 2021).
48. Martini, G.; Giechaskiel, B.; Dilara, P. Future European Emission Standards for Vehicles: The Importance of the UN-ECE Particle Measurement Programme. *Biomarkers* **2009**, *14*, 29–33. [CrossRef] [PubMed]
49. Giechaskiel, B.; Carriero, M.; Martini, G.; Krasenbrink, A.; Scheder, D. Calibration and Validation of Various Commercial Particle Number Measurement Systems. *SAE Int. J. Fuels Lubr.* **2009**, *2*, 512–530. [CrossRef]
50. Kupper, M.; Kraft, M.; Boies, A.; Bergmann, A. High-Temperature Condensation Particle Counter Using a Systematically Selected Dedicated Working Fluid for Automotive Applications. *Aerosol Sci. Technol.* **2020**, *54*, 381–395. [CrossRef]
51. Global Technical Regulation (GTR); Real-Driving Emissions (RDE). Informal Document GRPE-82-15. January 2021. Available online: <https://unece.org/sites/default/files/2021-01/GRPE-82-15e.pdf> (accessed on 7 December 2021).
52. UN Regulation on RDE 2021. Available online: <https://wiki.unece.org/display/trans/UNR+RDE> (accessed on 7 December 2021).
53. Giechaskiel, B.; Lähde, T.; Melas, A.D.; Valverde, V.; Clairotte, M. Uncertainty of Laboratory and Portable Solid Particle Number Systems for Regulatory Measurements of Vehicle Emissions. *Environ. Res.* **2021**, *197*, 111068. [CrossRef] [PubMed]

54. Park, G.; Mun, S.; Hong, H.; Chung, T.; Jung, S.; Kim, S.; Seo, S.; Kim, J.; Lee, J.; Kim, K.; et al. Characterization of Emission Factors Concerning Gasoline, LPG, and Diesel Vehicles via Transient Chassis-Dynamometer Tests. *Appl. Sci.* **2019**, *9*, 1573. [CrossRef]
55. Giechaskiel, B.; Melas, A.D.; Lähde, T.; Martini, G. Non-Volatile Particle Number Emission Measurements with Catalytic Strippers: A Review. *Vehicles* **2020**, *2*, 19. [CrossRef]
56. Vaaraslahti, K.; Virtanen, A.; Ristimäki, J.; Keskinen, J. Nucleation Mode Formation in Heavy-Duty Diesel Exhaust with and without a Particulate Filter. *Environ. Sci. Technol.* **2004**, *38*, 4884–4890. [CrossRef] [PubMed]
57. Lemmetty, M.; Rönkkö, T.; Virtanen, A.; Keskinen, J.; Pirjola, L. The Effect of Sulphur in Diesel Exhaust Aerosol: Models Compared with Measurements. *Aerosol Sci. Technol.* **2008**, *42*, 916–929. [CrossRef]
58. Lemmetty, M.; Vehkamäki, H.; Virtanen, A.; Kulmala, M.; Keskinen, J. Homogeneous Ternary $\text{H}_2\text{SO}_4\text{--NH}_3\text{--H}_2\text{O}$ Nucleation and Diesel Exhaust: A Classical Approach. *Aerosol Air Qual. Res.* **2007**, *7*, 489–499. [CrossRef]
59. Giechaskiel, B.; Bergmann, A. Validation of 14 Used, Re-Calibrated and New TSI 3790 Condensation Particle Counters According to the UN-ECE Regulation 83. *J. Aerosol Sci.* **2011**, *42*, 195–203. [CrossRef]
60. Giechaskiel, B.; Wang, X.; Horn, H.-G.; Spielvogel, J.; Gerhart, C.; Southgate, J.; Jing, L.; Kasper, M.; Drossinos, Y.; Krasenbrink, A. Calibration of Condensation Particle Counters for Legislated Vehicle Number Emission Measurements. *Aerosol Sci. Technol.* **2009**, *43*, 1164–1173. [CrossRef]
61. Sakurai, H.; Saito, K.; Taishi, T.; Koyama, T. Two Sources of Error in Determination of the Particle Concentration Reduction Factor of the Volatile Particle Remover Used in Legislated Vehicle Emission Measurement. *Meas. Sci. Technol.* **2011**, *22*, 085108. [CrossRef]
62. Otsuki, Y.; Nakamura, H.; Kojima, K.; Kondo, K.; Haruta, K. Investigation in Calibration Procedures for Portable and Stationary Solid Particle Number Measurement Systems. In *SAE Technical Papers*; 2019-01-1187; SAE International: Warrendale, PA, USA, 2019. [CrossRef]
63. Terres, A.; Giechaskiel, B.; Nowak, A.; Ebert, V. Calibration Uncertainty of 23nm Engine Exhaust Condensation Particle Counters with Soot Generators: A European Automotive Laboratory Comparison. *Emiss. Control Sci. Technol.* **2021**, *7*, 124–136. [CrossRef]
64. Giechaskiel, B.; Lähde, T.; Schwelberger, M.; Kleinbach, T.; Roske, H.; Teti, E.; van den Bos, T.; Neils, P.; Delacroix, C.; Jakobsson, T.; et al. Particle Number Measurements Directly from the Tailpipe for Type Approval of Heavy-Duty Engines. *Appl. Sci.* **2019**, *9*, 4418. [CrossRef]
65. Su, S.; Lv, T.; Lai, Y.; Mu, J.; Ge, Y.; Giechaskiel, B. Particulate Emissions of Heavy Duty Diesel Engines Measured from the Tailpipe and the Dilution Tunnel. *J. Aerosol Sci.* **2021**, *156*, 105799. [CrossRef]
66. Krajinska, A. Opportunities for Future Tailpipe Emissions Regulation of Light-Duty Vehicles within the European Union. In *Transportation Air Pollutants*; Brewer, T., Ed.; Springer: Cham, Switzerland, 2021; pp. 55–69. ISBN 978-3-030-59690-3.
67. ICCT. *ICCT's Comments and Technical Recommendations on Future Euro 7/VII Emission Standards*; ICCT: Berlin, Germany, 2021; Available online: <https://theicct.org/sites/default/files/eu-commission-euro-7-and-VI-may2021.pdf> (accessed on 7 December 2021).
68. AGVES. *Advisory Group on Vehicle Emission Standards (AGVES)*; AGVES: Brussels, Belgium, 2019; Available online: <https://circabc.europa.eu/faces/jsp/extension/wai/navigation/container.jsp> (accessed on 7 December 2021).
69. Kontses, A.; Triantafyllopoulos, G.; Ntziachristos, L.; Samaras, Z. Particle Number (PN) Emissions from Gasoline, Diesel, LPG, CNG and Hybrid-Electric Light-Duty Vehicles under Real-World Driving Conditions. *Atmos. Environ.* **2020**, *222*, 117126. [CrossRef]
70. Dimaratos, A.; Toumasatos, Z.; Triantafyllopoulos, G.; Kontses, A.; Samaras, Z. Real-World Gaseous and Particle Emissions of a Bi-Fuel Gasoline/CNG Euro 6 Passenger Car. *Transp. Res. Part D Transp. Environ.* **2020**, *82*, 102307. [CrossRef]
71. Toumasatos, Z.; Kontses, A.; Doulgeris, S.; Samaras, Z.; Ntziachristos, L. Particle Emissions Measurements on CNG Vehicles Focusing on Sub-23nm. *Aerosol Sci. Technol.* **2021**, *55*, 182–193. [CrossRef]
72. Liu, G.; Liu, W.; He, Y.; Gong, J.; Li, Q. Research on Influence of Exhaust Characteristics and Control Strategy to DOC-Assisted Active Regeneration of DPF. *Processes* **2021**, *9*, 1403. [CrossRef]
73. Tong, Y.; Tan, J.; Meng, Z.; Chen, Z.; Tan, L. Experimental Investigation on the DPF High-Temperature Filtration Performance under Different Particle Loadings and Particle Deposition Distributions. *Processes* **2021**, *9*, 1465. [CrossRef]
74. Liu, S.; Lü, M. Fault Diagnosis of the Blocking Diesel Particulate Filter Based on Spectral Analysis. *Processes* **2019**, *7*, 943. [CrossRef]
75. Orihuela, M.P.; Haralampous, O.; Chacartegui, R.; Torres García, M.; Martínez-Fernández, J. Numerical Simulation of a Wall-Flow Particulate Filter Made of Biomimetic Silicon Carbide Able to Fit Different Fuel/Biofuel Inputs. *Processes* **2019**, *7*, 945. [CrossRef]
76. Valverde, V.; Giechaskiel, B. Assessment of Gaseous and Particulate Emissions of a Euro 6d-Temp Diesel Vehicle Driven >1300 km Including Six Diesel Particulate Filter Regenerations. *Atmosphere* **2020**, *11*, 645. [CrossRef]
77. Giechaskiel, B. Particle Number Emissions of a Diesel Vehicle during and between Regeneration Events. *Catalysts* **2020**, *10*, 587. [CrossRef]
78. Amanatidis, S.; Ntziachristos, L.; Giechaskiel, B.; Bergmann, A.; Samaras, Z. Impact of Selective Catalytic Reduction on Exhaust Particle Formation over Excess Ammonia Events. *Environ. Sci. Technol.* **2014**, *48*, 11527–11534. [CrossRef] [PubMed]
79. Mamakos, A.; Schwelberger, M.; Fierz, M.; Giechaskiel, B. Effect of Selective Catalytic Reduction on Exhaust Nonvolatile Particle Emissions of Euro VI Heavy-Duty Compression Ignition Vehicles. *Aerosol Sci. Technol.* **2019**, *53*, 898–910. [CrossRef]
80. Suarez-Bertoa, R.; Astorga, C. Impact of Cold Temperature on Euro 6 Passenger Car Emissions. *Environ. Pollut.* **2018**, *234*, 318–329. [CrossRef]

81. Siedlecki, M.; Merkisz, J.; Dobrzyński, M.; Kubiak, K. Impact of the Use of Comfort Devices on the Exhaust Toxic Compounds from a Modern PC Car with Spark Ignition Engine. *Combust. Engines* **2021**. [[CrossRef](#)]
82. Giechaskiel, B.; Valverde, V.; Kontses, A.; Melas, A.; Martini, G.; Balazs, A.; Andersson, J.; Samaras, Z.; Dilara, P. Particle Number Emissions of a Euro 6d-Temp Gasoline Vehicle under Extreme Temperatures and Driving Conditions. *Catalysts* **2021**, *11*, 607. [[CrossRef](#)]
83. Valverde-Morales, V.; Clairotte, M.; Pavlovic, J.; Giechaskiel, B.; Bonnel, P. On-Road Emissions of Euro 6d-TEMP Vehicles: Consequences of the Entry into Force of the RDE Regulation in Europe. In *SAE Technical Papers*; 2020-01-2219; SAE International: Warrendale, PA, USA, 2020. [[CrossRef](#)]
84. Giechaskiel, B.; Zardini, A.; Martini, G. Particle Emission Measurements from L-Category Vehicles. *SAE Int. J. Engines* **2015**, *8*, 2322–2337. [[CrossRef](#)]
85. Kontses, A.; Ntziachristos, L.; Zardini, A.A.; Papadopoulos, G.; Giechaskiel, B. Particulate Emissions from L-Category Vehicles towards Euro 5. *Environ. Res.* **2020**, *182*, 109071. [[CrossRef](#)]
86. Giechaskiel, B. Differences between Tailpipe and Dilution Tunnel Sub-23 nm Nonvolatile (Solid) Particle Number Measurements. *Aerosol Sci. Technol.* **2019**, *53*, 1012–1022. [[CrossRef](#)]
87. Giechaskiel, B. Effect of Sampling Conditions on the Sub-23 nm Nonvolatile Particle Emissions Measurements of a Moped. *Appl. Sci.* **2019**, *9*, 3112. [[CrossRef](#)]
88. Bielaczyc, P.; Honkisz, W.; Woodburn, J.; Szczotka, A.; Forloni, F.; Lesueur, D.; Giechaskiel, B. Inter-Comparison of Particle and Gaseous Pollutant Emissions of a Euro 4 Motorcycle at Two Laboratories. *Energies* **2021**, *14*, 8101. [[CrossRef](#)]
89. Giechaskiel, B.; Melas, A. Emissions of a Euro 5 Motorcycle over the World Harmonized Motorcycle Test Cycle (WMTC). *Combust. Engines* **2021**, *185*, 21–25. [[CrossRef](#)]
90. Burtscher, H.; Lutz, T.; Mayer, A. A New Periodic Technical Inspection for Particle Emissions of Vehicles. *Emiss. Control Sci. Technol.* **2019**, *5*, 279–287. [[CrossRef](#)]
91. Giechaskiel, B.; Lähde, T.; Suarez-Bertoa, R.; Valverde, V.; Clairotte, M. Comparisons of Laboratory and On-Road Type-Approval Cycles with Idling Emissions. Implications for Periodical Technical Inspection (PTI) Sensors. *Sensors* **2020**, *20*, 5790. [[CrossRef](#)] [[PubMed](#)]
92. Grigoratos, T.; Martini, G. Brake Wear Particle Emissions: A Review. *Environ. Sci. Pollut. Res.* **2015**, *22*, 2491–2504. [[CrossRef](#)]
93. Harrison, R.M.; Allan, J.; Carruthers, D.; Heal, M.R.; Lewis, A.C.; Marner, B.; Murrells, T.; Williams, A. Non-Exhaust Vehicle Emissions of Particulate Matter and VOC from Road Traffic: A Review. *Atmos. Environ.* **2021**, *262*, 118592. [[CrossRef](#)]
94. Schriebl, M.A.; Bergmann, A.; Fierz, M. Design Principles for Sensing Particle Number Concentration and Mean Particle Size with Unipolar Diffusion Charging. *IEEE Sens. J.* **2019**, *19*, 1392–1399. [[CrossRef](#)]
95. Schriebl, M.A.; Nishida, R.T.; Knoll, M.; Boies, A.M.; Bergmann, A. Characterization of Particle Number Counters Based on Pulsed-Mode Diffusion Charging. *Aerosol Sci. Technol.* **2020**, *54*, 772–789. [[CrossRef](#)]