RESEARCH



Nanoparticle Counting for PTI: The Dirty Tail Paradigm

A Pragmatic Proposal to Strongly Reduce Urban PN Pollution from Combustion Engine Fleets

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Abstract

Using solid particle number (PN) measurements in the European Periodic Technical Inspection (PTI) of diesel engines equipped with particulate filters was proposed by VERT in 2016 during the Dieselgate Hearing of the Federal Republic of Germany. An international working group developed the standards and instruments for this method over 3 years under the leadership of TNO and VERT, which were next implemented in four countries, Belgium, the Netherlands, Germany, and Switzerland, starting in 2022. PN measurement is now state of the art, enabling rapid and reliable detection of possible failures in particulate filters and the need for their immediate restoration. This paper expands on that successful experience, recommending that PN counting be used for control of PN emissions from all vehicles during PTI. It reviews results from a number of earlier studies on "high emitters" and shows some new data sets for gasoline engines. Five large vehicle fleets, diesel and gasoline, heavy-duty engines (HDE), light-duty vehicles (LDV), and non-road mobile machinery (NRMM), with and without emission aftertreatment were analyzed. It was observed that, while most vehicles in working fleets are clean (i.e., meet or often are far are below their corresponding emission limits), every fleet, however, contains some high emitters, about 4-8% of the fleet hereby termed "dirty tail" — diesel as well as gasoline engines. This small fraction dominates the PN emission of the entire fleet and may increase the overall PN emission of its corresponding fleet by more than tenfold over the level of the compliant vehicles! Experience indicates that PN emission may be a strong indicator of many different deteriorations in a combustion engine and thus can be used as a highly sensitive diagnostic signal to detect various engine or emission faults quickly and reliably. This is a new understanding of emission control of vehicle fleets: not by regulations for new vehicles only which apply for all vehicles but by selecting the high emitters and consequently repair or replace these relatively few vehicles to the extent desired in terms of emissions policy. Most countries have already implemented strong periodic technical inspection systems. We suggest to expand such tests by additionally measuring the particle number concentration in the exhaust gas of all vehicles for just 1 min, thereby detecting the high emitters. With consistent annual monitoring, this procedure will reduce urban particle pollution from combustion engines to one-tenth or lower, a significant contribution to reducing local health risks.

Keywords Periodical technical inspection · Superpolluter · Emissions from combustion engines

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1 Introduction

Ensuring lifetime emission quality of all vehicles in the entire vehicle fleet, an important element of regulations of most countries, requires a hierarchy of controls:

- Homologation (type approval) guarantees that new generations of vehicles comply with current legislation, including foreseeable deterioration due to thermal or chemical ageing.
- Monitoring the conformity of production (COP) guarantees the uniformity of the technology produced, whereby

individual quality fluctuations in parts or processes cannot be avoided and can accumulate.

• In-service conformity monitoring (ISC) (or surveillance monitoring) monitors ageing and systematic deterioration effects, but is limited to testing very few units for cost reasons. A statistically meaningful result cannot be derived from this due to the small number of samples.

Today, these three principles are generally applied for the road vehicle fleet. However, these measures only cover *systematic* deterioration effects that are caused by the technology used and the established production quality. Re-introduction of PTI including a PN measurement also clearly showed that the expectation that OBD on-board diagnostics, which is required in modern vehicles, will find all faults is not realistic (see, for example, [1, 2]).

The abundant number of possible statistical failures, negligent maintenance and deliberate manipulation, can have a much greater impact on the emissions of individual vehicles than foreseeable systematic deterioration factored into the assessment [2]. Already Kurinam and Schmidt-Ott [3] found that a small fraction of cars, they called them "superpolluters," are responsible for a large fraction of the fleet emissions. As the data presented in the following clearly shows, few vehicles emit up to10,000 times more ultrafine particles than the best ones and more than 1000 times more than the majority of compliant vehicles. The average value of the fleet is thus raised far above the legal limits. COP and ISC do not find this, nor does OBD apparently, neither it is detectable in traffic by the police and if the entire vehicle fleet is not checked for such high emitters, we could observe very high levels of air pollution despite strict emission limits, which are complied with by the majority. Further tightening of limits would then be ineffective. As the presented data show, this is the realistic scenario. They also show that this actually applies to all fleets, but until recently has so far been ignored by emissions policy. However, the problem can be solved in a simple way if we generally introduce a highly sensitive criterion with the particle number concentration, which can be checked for all vehicles during the normal PTI in a very short time and at low cost. If we assume that the "dirty tail paradigm" applies to all fleets with internal combustion engines, a very likely assumption as we will show, this opens up a completely new and very simple way of cleaning the urban particle pollution resulting from traffic in the air we breathe by a factor of five to ten in a very short time.

2 Solution Approach

In contrast to widespread political approaches for so-called open filters or bypass filters for exhaust gases [4], VERT has demanded "best available technology" from the outset, as is the case for any carcinogenic air pollutant [5]. With the demand for filtration efficiencies > 98% from a particle size of 10 nm, the bar was set very high as early as 1998 and consistently maintained. VERT has thus taken on a leading role and advanced the technology with this strict requirement, making it the only independent certification body to register over 70 systems with an average filtration efficiency of 98.8% [6]. In practice, these filters are reducing particle emissions in the entire alveolar size range by at least two orders of magnitude, the best even by three to four orders of magnitude [7].

Conversely, however, every filter failure massively increases the emissions of a vehicle fleet equipped with filters. The question therefore arose as to how this quality can be ensured in practice. After the EU recommended in 2012 [8] that periodic emissions testing be dispensed with altogether and that this control function be left entirely to the manufacturer as part of the electronic on-board diagnostics OBD, the quality crisis was pre-programmed. It culminated with the VW scandal, when it became apparent that even massive manipulations in the exhaust technology were not detected by the OBD — the MIL, the malfunction indicator light, did not light up.

This led to a hearing of the German Bundestag on September 23, 2016 (5.PUH), at which the VERT representative took the initiative with the proposal to reintroduce the periodic exhaust gas control and to supplement it with a measurement of the number concentration of solid particles from 23 nm [9]. The proposal was soon adopted in the German Road Traffic Licensing Regulations (STVZO) in 2017 and the introduction of a number measurement was announced for January 1, 2021 [10], but standards for the measuring instruments and a regulation for carrying out the PTI test were still missing. The experience available at VERT suggested carrying out these measurements at low engine idle and developing measuring devices for this purpose, which should enable general introduction in vehicle workshops at a cost of < 8000 euros [11]

The "NPTI" engineering task force was founded together with TNO in December 2016 with this vision of a specification. The database was expanded through measurements in Belgium, the Netherlands, Spain, Mexico, Switzerland, and the EU JRC, and the standards for the test procedure and the measuring instruments were developed with the help of the Dutch Metrology Institute NMI [12, 13]. JRC investigated the correlation of the measurements in idling mode to the WLTC-test cycle used for type approval [14]. The instrument development was taken up by over 20 companies so that legal implementation could be started in 2019. [15]. The result of this work is presented in [16, 17].

3 Experimental Findings

The following quality controls of large vehicle fleets show a uniform picture:



Fig.1 Emissions from construction machinery sorted according to the number concentration of particles; the dirty tail becomes clear [21]

3.1 Construction Machinery

Field controls were already introduced for construction machinery in Switzerland with the Construction Directive in 2002 [18]. As the available CPC devices were not suitable for field use, Matter Engineering AG, in cooperation with VERT and the ETH, developed a measuring device for this purpose as early as 1997, the NanoMet [19], which recorded the particle concentration with two sensors, namely DC according to the principle of diffusion charging and PAS, the photoelectric aerosol sensor. DC prevailed, in combination with dilution and volatile removal by heating, catalysis or absorption. The first official Swiss guideline was published in 2012 [20], whereby the measurement was carried out in the upper idle speed as on the opacity measurement, which was replaced. From 2012, a CPC device from TSI, the NPET, which had already been developed in accordance with this directive, came onto the market. A fleet of 107 construction machines with running times of up to 18 years and up to 12,000 operating hours delivered the results, shown in Figs. 1 and 2. Figure 1 shows the absolute PN emission as function of the relative number of vehicles; Fig. 2 shows the cumulative emission, which is calculated by integrating the data in Fig. 1 and normalizing the result to 100% for the sum of all emissions.

A typical picture emerges: Relatively few vehicles are responsible for most of the emissions of the entire fleet. Eighty percent of PN emissions come from 10% of the machines and 95% from 20%. In [21], the emissions are also shown as function of engine age, operating hours, and rated power. These parameters have a minor influence.



Fig. 2 Cumulative impact on the emissions of the entire fleet [21]

3.2 400 Public Transport Buses with DPF in Santiago, the CH-CALAC project 2014/2015

As part of Switzerland's cooperation with Chile, public transport buses in Santiago were retrofitted with VERT-certified particle filters from 2004 [22]. In 2014, when almost the entire bus fleet was equipped, a broad roadside quality check was carried out on 400 vehicles. The vehicles were randomly selected for this purpose, taken out of traffic by the police for one minute and the measurement had to be carried out within this time — a demanding requirement for an ad hoc roadside check. The opportunity was taken to compare with the opacity measurement, which was still popular at the time, with the particle count measurement. As the NPET measuring device selected for this purpose is highly dynamic, three measurement modes could be compared within this short period of time, namely stationary low idle speed, free acceleration up to the stall speed and stationary measurement in upper idle speed.

The following image, Fig. 3, shows the test sequence with the continuously recorded particle concentration from low idle speed via free acceleration to high idle speed (speed control) in three repetitions, for a total of 60 s. It led to the findings that a dynamic measurement (free acceleration) is not necessary, that the opacity measurement was not capable of detecting errors, and, furthermore, that the measurement at low idle provides well reproducible and stable measured values [23].

The results obtained at low idle were sorted into 16 emission classes, as shown in Table 1 and Fig. 4. The height of the columns corresponds to the number of buses per emission class. This resulted in a clearly bimodal distribution



Fig. 3 Number emission PN in the triple repetition of the combined test cycle with the vehicle stationary with measurement at low idle, at cutoff speed and transient measurement under free acceleration, measured within one minute with an NPET measuring device from TSI [23]

Table 1 Concentration range in classes 1–16 1	Class	Range [cm ⁻³]
	1	$1.0\ 10^2$ – $2.2\ 10^2$
	2	$2.2 \ 10^2 - 4.7 \ 10^2$
	3	4.7 10 ² -1.0 10 ³
	4	$1.0\ 10^3$ - $2.2\ 10^3$
	5	$2.2\ 10^3$ - $4.7\ 10^3$
	6	$4.7 \ 10^3 - 1.0 \ 10^4$
	7	$1.0\ 10^4$ – $2.2\ 10^4$
	8	$2.2 \ 10^4$ - $4.7 \ 10^4$
	9	$4.7 \ 10^4 - 1.0 \ 10^5$
	10	$1.0\ 10^{5}$ - $2.2\ 10^{5}$
	11	$2.2\ 10^{5}$ - $4.7\ 10^{5}$
	12	4.7 10 ⁵ -1.0 10 ⁶
	13	$1.0\ 10^{6}$ - $2.2\ 10^{6}$
	14	$2.2\ 10^{6}$ - $4.7\ 10^{6}$
	15`	$4.7 \ 10^{6} - 1.0 \ 10^{7}$
	16	$1.0\ 10^7 - 2.2\ 10^7$

with an enormous spread of PN emissions over four orders of magnitude and the possibility of forming a sensible pass/fail limit value for this technical level, here at 220,000 cm⁻³. Ten percent of vehicles cause 80% of emissions (Fig. 5).

It became clear that relatively few vehicles in the high emission classes are responsible for the total PN emissions of the fleet. Just 1 min of measurement, even at the roadside, was enough to identify the faulty vehicles and assign them for repair.

Since in Chile, as in many countries, a periodic exhaust gas check is standard — every 6 months for buses — there is not even any additional cost for this quality control. The PN measurement simply replaces the opacity measurement. The measurement at low idle even simplifies the effort compared to free acceleration, and the police have been given a means of checking the quality of emissions in traffic.

3.3 1009 Diesel Cars with OE Particle Filters in Zurich 2017/2018

In May 2000, Peugeot launched the first vehicle equipped with a particle filter as standard and achieved great sales success. Several other OEMs followed, even though the PN regulation was not yet in force in the EU. In Switzerland, where particle filters had already been introduced for construction trucks and construction machinery as well as for public transport buses, ships, and locomotives, the passenger car fleet with filters also grew even before Euro 5.

B. Gloor (AWEL, Zurich) [24] examined a total of 1090 diesel cars with DPF during the normal PTI in the largest motor vehicle inspection in the canton of Zurich in 2017 as part of a research program initiated by him. Emission measurement as part of the PTI had been abandoned in Switzerland and throughout Europe in 2013 in favor of the electronic self-monitoring OBD following 2014/45 of the EU in 2012. He used various instruments, including DC and CPC. The particle number concentration was measured under low idle conditions. In 2011, with Euro 5b, the number measurement was introduced by the EU during type approval testing, making the particle filter the state of the art in diesel passenger car technology, and the number of registrations with filters increased accordingly. As the PTI obligation in Switzerland only begins after 5 years of operation, the figures fall thereafter; only special vehicles such as taxies are checked earlier.

Of the vehicles, 12.5% equipped with filters in the 2011 enrolment year exceeded the limit value of $250,000 \text{ cm}^{-3}$ applied here, which was attributed to cracks, damage, manipulation of the filter, or the system. However, the type of damage in these failures was not analyzed further. The following graph, Fig. 6, shows the typical behavior: Less than 10% of vehicles are responsible for the majority of the







Fig. 5 The normalized representation shows the influence of a comparatively small number of vehicles on the total emissions of the fleet

fleet's particle emissions. Figure 7 shows the results ordered by the yeas of first registration. If it is so easy to identify these faults, it would be a simple matter to massively reduce particle emissions by replacing or repairing these relatively few vehicles. The measurements were repeated a year later and the failure rate was slightly lower, presumably because the OEM's DPF technology had made progress in the meantime.

Measurements of this type were also carried out in Belgium, the Netherlands, Germany, and Spain and presented at the VERT Forums 2017 to 2024; in total, more than 5000 vehicles were tested to substantiate the argument for the introduction of PN-PTI [25].

All of these cases involved diesel engines with DPF.

The following investigations are concerned with gasoline vehicles without filters, so any excessive particle emissions can no longer be attributed to filter failure; they are to be attributed to other sources of PN in the engine system.



Fig. 6 Cumulative contribution to PN emissions from high-emitting vehicles (with DPF damage) in the diesel car fleet with DPF in Zurich [26]

3.4 Petrol LDV in Mexico City

Until 2017, particulate limits existed worldwide only for diesel engines, initially based on opacity and later on particulate mass, as older diesel engines often emitted visible soot. The fact that petrol engines can also emit high concentrations of ultrafine particles only became apparent with the introduction of direct injection. In reality, gasoline engines always emitted ultrafine particles, often less than diesel, but above all significantly smaller and therefore even less visible and more dangerous from the point of view of translocation in the organism. The PAH content of these particles and thus their toxicity potential is also massively higher than that of diesel [27].

In a project on reducing particle emissions in Mexico City in 2017, VERT used the DiSCmini particle number measuring device for ambient air from Matter Engineering **Fig. 7** Distribution of the number of cars complying with the limit value of 250,000 cm⁻³ (green) and those exceeding the limit (red) as a function of vehicle operating time



AG, which was already available at the time and able to also indicate particle size to show that, in addition to HDVs and NRMMs, primarily petrol vehicles pollute the air in this city with ultrafine particles. This statement most probably applies to all non-European cities, as, in most countries outside Europe, gasoline vehicles predominate among passenger cars.

The air pollution control authority SEDEMA took up this observation and installed NanoMet3 particle measuring devices from Matter/Testo, which complies with the EU PMP protocol, in all 60 PTI measuring stations and carried out particle measurements on over 400,000 vehicles from 2018. In this case, the measurements were even carried out on the chassis dynamometer in two short test cycles, namely in the US-ASM 5024 and followed by US-ASM 2540 cycles, i.e., at 25 and 40 km/h and corresponding load.

The data were analyzed with the support of the EU/JRC and are available as an EU-publication [28].

This fleet in Mexico City does not contain any filters, but the majority have three-way catalytic converters with lambda control. What we see here, in contrast to the previous examples, are not failures of particle filters but other failures of different sources, which also lead to the typical dirty tail behavior due to their random nature: 12% of the vehicles are responsible for 85%, 2% for 62% of particle emissions, and reach concentrations of up to 100 million particles cm⁻³. Older vintages show a broader influence, which seems plausible as a result of increasing wear and tear (Figs. 8 and 9).

These observations thus suggest that dirty tail behavior due to the accumulation of failures exists in general and that the PN measurement at vehicle standstill and engine idling



Fig. 8 PN emission distribution for the total fleet of 400,000 vehicles in Mexico City tested during PTI, sorted by year of manufacture. Peak PN concentration reaches 10^8 cm^{-3}

in the PTI offers the possibility of tracking down many failures in this simple way.

3.5 Gasoline LD Vehicles in Switzerland 2023/2024

As part of the EU Horizon project AeroSolfd 2022–25 [29], VERT investigates a fleet of 1000 gasoline vehicles using a DC-PN-Counter from AVL DiTEST developed and certified for the NPTI project with the primary objective of PTI control of diesel cars with DPF. The vehicle selection is random and is carried out following the normal PTI test in the official test center of the TCS-Section Biel/Bienne Seeland, i.e., all vehicles are included as they appear for



Fig. 9 PTI test in a test center in Mexico City with dynamic recording of the particle number concentration by NanoMet3 in the ASM 2540 and ASM 5024 dyno test at 24 km/ and 40 km/h

PTI, most of which are older than 5 years, a few also with GPF. This study is intended to provide information on the PN emissions of the current fleet of petrol vehicles in Switzerland.

Following preliminary investigations by NPTI partners such as JRC, TÜV and AVL DiTEST, the test protocol was defined in such a way that measurements were carried out with the vehicle at a standstill at an increased idling speed of 2000 rpm, without loading the engine and as a variant with loading by switching on the air conditioning system and additional electrical consumers.

The PN emissions of many petrol engines are quite low, in the range of $30,000-50,000 \text{ cm}^{-3}$, but even in this modern fleet, which is certainly well maintained according to Swiss criteria, there are high emitters that exceed 10^7 cm^{-3} and thus even the upper measuring limit of the devices used today (Figs. 10 and 11). Incidentally, the measuring devices used here are already prepared for these petrol engines by separating the high water content of the exhaust gas of petrol engines, and they have proven with their NPTI certification that they meet the "volatile particle removal" condition. This means that they are definitely solid particles.

It is interesting to note from the statistical data that although the mean values for the younger emission classes are significantly lower, high-emitting vehicles with particle concentrations at the measurement limit of the instrument with 10 million particles cm⁻³ occur in all Euro classes (Figs. 12 and 13).

Since the soot particles from gasoline engines not only have higher content of heavy metals than diesel particles [30] and also higher concentrations of highly toxic and carcinogenic PAHs [31, 32], it seems urgently necessary to set strict limit values and the failure criteria for gasoline engines.



Fig. 10 Dirty tail of the Swiss gasoline fleet



Fig. 11 Cumulative contribution to fleet emissions

3.6 A Summary of the Fleet Findings

The following parameters can be regarded as typical parameters for the evaluation of PN emission behavior under the influence of dirty tail effects.

- Average PN emission pf the fleet
- Average PN emission of all "clean" vehicles below the pass/fail limit value
- Maximum value
- Minimum value
- Median
- Percentage below and above the pass/fail criterion



Fig. 12 The composition of this fleet. As new cars have the first PTI only after 5 years, the last 5 years are missing, and only a few vehicles with GPF are included



Fig. 13 Although all emission classes show a significant reduction in gaseous emissions such as NOx, THC, and CO over this period, the values for PN remain high with pronounced maximum values

The following graph, Fig. 14, compares the first three parameters for all five fleets.

The result is remarkably consistent and proves that the total emissions of all these five fleets can be reduced by more

than an order of magnitude by applying the dirty tail fleet emission cleaning methodology.

This is amazing, considering that very different reasons are responsible for this behavior. In the case of diesel engines, the main problem seemed to be DPF failures, but almost all of the gasoline engines have no filter. The high emissions therefore must be due to some kind of engine malfunction as for example incorrect operation of the λ -control.

4 Why PN Was Chosen as a Quality Criterion

4.1 PN Is the Reference Value for the Toxic Health Effects of Automobile Exhaust Gases

Most epidemiological studies refer to PM10, thus underestimating the effects of ultrafine particles according to their size and substance and are therefore almost worthless from the point of view of the risk posed by ultrafine particles. The statements are only marginally better if PM2.5 is chosen as the pollutant [33, 34], but even then, the mass is determined by large particles and the substance is undefined. The carcinogenicity was only recognized [35] and confirmed by the WHO when studies were included that only referred to ultrafine particles from diesel engines [36]. The aim must therefore be to relate the health risks to the actual toxic substance, i.e., the number of insoluble particles with a mobility diameter < 500 nm [37] and their contents, which are introduced into the organism in the sense of the Trojan Horse effect [8] and translocated anywhere inclusive into the brain and fetus. The particles emitted by combustion engines belong to this class, and only a few such particles come from other sources.

One of the latest studies by the Harvard School of Health [38] takes this fact into account by focusing exclusively on



Fig. 14 The three most important parameters of fleet emissions: maximum value, average of compliant vehicles below the limit value of $250,000 \text{ cm}^{-3}$, and overall average with the influence of the dirty tail traffic-related particles. It concludes that these particles are responsible for > 95% of adult mortality from traffic-related air pollution in the 6.2 million per year (children under 14 were not included in this study based on insurance data in the USA), mainly from heart attacks, strokes, and lung cancer. These particles are produced by combustion and nucleation from the gas phase [39], and the main source in urban air is combustion engines, the No.1 Toxic Air Contaminant according to the US EPA [40]. The total mass PM (mg/m^3) , mg/kWh) of these tiny particles is extremely low and is often below the detection limit of gravimetric detection in modern engines, but the number concentration of ultrafine particles PN (1/cm³, 1/kWh), is high and can be measured very accurately even in clean engine exhaust gas at low idle speeds. Counting the particles also takes into account the heavy metal content and the accumulated PAH and thus the actual toxicity. Nitrogen oxide emissions, on the other hand, which are also frequently cited in this context, are not carcinogenic and do not appear at all in this epidemiological classification of causes of mortality. According to [38], ozone, which is partly formed from nitrogen oxides, has an influence of < 5%on overall mortality due to air pollution from traffic.

Various studies show that the best correlation to health effects is achieved with the particle surface [41], which is probably related to the "Trojan horse effect," i.e., the hypothesis that the harmful substances are adsorbed on the particle surface and carried in the organism. Another useful metric would be the concentration of elemental carbon, which is used in occupational medicine. This parameter would have the advantage that it is a conservation parameter in which emission and immission values can be directly compared. However, as the number concentration has already become established as a metric for emissions from combustion engines for pragmatic reasons, it makes sense to stick with it for periodic exhaust gas monitoring as well, otherwise comparability would be lost.

4.2 The Measuring Sensitivity of PN Counting Is Much Greater Than That of the Gravimetric Determination of the Mass PM

Of the common PN counting measuring devices, CPCs are able to count particles individually, i.e., down to a few particles per cubic centimeter. For DC devices, the lower limit is a few hundred particles per cubic centimeter, i.e., around 10^8 particles per m³ of exhaust gas [42]. With the mass of one femtogram (10^{-15} g) per particle of 100 nm, this corresponds to a total mass of 10^{-7} g, i.e., 0.1 µg. The detection limit for PM is currently specified as 0.1 mg/m³ for the best systems [43], so measuring the number of particles is around 1000 times more sensitive than a gravimetric measurement, even with the inexpensive NPTI devices. In addition, it is simpler, faster, and even transiently meaningful up to 10 Hz.

Thanks to the general introduction of NPTI, such devices are now also available in a wide range and suitable for use in workshops.

4.3 PN Is a Very Sensitive Signal for Many Potential Particle Creating Processes in the Engines [44]

Just a few examples:

- Injection problems (pressure, time, direction) increasingly generate PN due to the formation of zones of imperfect combustion with a lack of oxygen.
- Turbocharger seal ring wear generates increased PN through the increased lubricating oil input into the combustion process or directly into the exhaust gas.
- Valve leakage generates increased PN by intensifying the valve overlap effect and thus shifting the mixture towards "rich".
- Experience has shown that altitude compensation by increasing the turbocharger speed results in increased wear of bearings and sealing rings and thus more lubricating oil entering the exhaust gas.
- Fuel problems influence PN formation in many ways: insufficient filtration, ash-forming agents and sulfur content are particularly problematic.
- EGR problems (sooting of lines and valves, cooling) are heavily involved in PN formation, as the engine reacts immediately to the corresponding change in the mixture parameters.
- DPF problems (hairline cracks, preload mat) reduce the particle separation rate surprisingly quickly and strongly.
- Piston, liner or piston ring wear generate increased PN, not only due to the wear particles that can vaporize and renucleate in the combustion process, but also by increasing the lubricating oil input into the combustion process.
- Lubricating oil composition is due to the deliberate addition of metallic additives (P, Zn) and sulfur is a decisive factor for the increased PN formation and for the shortening of the DPF service life.
- The so-called blow-by system, i.e. the recirculation of the lubricating oil mist from the crankcase via droplet separation into the air intake, plays a key role. An increase in the lubricating oil level in the crankcase is sufficient to intensify this process.
- Deterioration of the intake filter due to dust overload or leaks reduces the intake air throughput for combustion, thereby promoting PN formation and leading to increased wear.
- Deterioration of the NO₂ formation, which is important for regeneration, due to ageing of the DOC coating in the diesel engine can be the reason for early damage to the filter matrix.
- Deterioration of the TWC in gasoline engines due to ageing of the coating primarily increases the gaseous emis-

sions NOx and THC, but the higher-boiling HCs also can generate nanoparticles during cooling.

- Failures in the lambda control naturally produce nanoparticles when drifting into the rich range.
- With SCR, it is known that urea crystallites and ammonium sulfate particles can form. If these are detected early by a PN measurement, clogging of the SCR catalytic converter can be avoided.
- Changes of combustion strategies in some types of modern engines, which were developed with no restrictions, or considerations of PN emissions.

Even more impressive is the enormous range of particle concentrations in these cases: A diesel engine with a good DPF has PN emissions below 1000 cm^{-3} , but in the case of damage to the filter matrix, this number can rise to more than 10^7 cm^{-3} , and the spread over four order of magnitudes can be detected quite accurately with reliable and inexpensive instruments. While these are easy to understand for particle filter damage, the effect in many other, more complex failure modes can be equally large but has not yet been adequately described. For example, a good gasoline engine, even without a GPF, can have PN emissions below 1000 cm⁻³, but some of these engines [28], even when equipped with a TWC, can emit up to 10^8 cm^{-3} as we have seen with the Mexican fleet.

The PN measurement as done in PTI tells us only that there is damage and how extensive it is, but it does not reveal the technical cause. However, this is decisive to know for the workshop. The methodology of fault diagnostics is therefore an important task for the future in order to provide workshops with rules on how to proceed in order to find and rectify the actual damage. As the following suggestions show this requires additional measurements, which preferably are done in workshops, commissioned by the manufacturer which then may lead to guidelines how to deal with the problem. Some suggestions include measuring PN upstream of the filter, checking the gaseous composition of the exhaust gas, determining at least the oxygen content and CO, checking the EGR function, and lubricating oil content of the particulate matter. This already allows some faults to be narrowed down. The usual checks, such as engine compression, blow-by pressure, boost pressure behavior, checking all system filters, and deposits in the exhaust pipe, together with the fault memory analysis, will probably quickly lead to a clear diagnosis.

5 The PN Measurement Technology

The specifications proposed originally by VERT for the diesel measuring equipment where [11, 16]:

• Measurement at low idle

- Efficiency $100\% \pm 25\%$ at 55 nm
- Efficiency $50\% \pm 25\%$ at 23 nm
- Deposition efficiency of volatile components > 90% for 30 nm tetracontane (a high boiling oil C40H82) particles with a number concentration < 10⁵ cm.⁻³
- Pass/fail at 50,000 cm.⁻³

On this basis, the VERT-NPTI project was launched with TNO in December 2016.

After 10 meetings of this working group, the following specifications have now been included in the Dutch legislation 2022 for NPTI [12, 13, 45].

- Detection efficiency Particle size Detection efficiency. 23 nm±5%20-60% 50 nm±5%60-130% 80 nm±5%70-130%
- Measurement at low idle.
- Separation efficiency of volatile components > 95% for 30 nm tetracontane particles with a number concentration $< 10^5$ cm⁻³.
- Measuring range: $5000-5 \times 10^6$ cm⁻³.
- Pass/fail at 10^6 cm^{-3} .

Similar, but unfortunately not identical requirements exist for Germany from 2023 [46] and for Switzerland in 2023 [47], where they have been in place for construction machinery since 2012 [20] and for ships on Swiss lakes since 2017.

A summary of the specifications can be found in [48].

Two methods are currently used for measurement, namely condensation particle counters (CPC) and devices based on the electrical charging of particles through the attachment of ions (diffusion charging, DC devices). With CPC, the particles in a supersaturated vapor grow to a size that allows them to be optically detected and counted [49]. This makes the measurement very sensitive and even the smallest concentrations can be easily measured. Measurement uncertainties occur with very small particles, where the probability that they will grow is no longer 100% and at high concentrations due to agglomeration and deviations in the flow rate. Because of the agglomeration risk, dilution must always be used with these devices in order to achieve a sufficiently large measuring range.

With DC devices, an electrical current is measured from which the number is determined assuming the probability of charging depending on the particle size. As it is not possible to measure arbitrarily small currents, the detection limit is a few hundred particles per cubic centimeter, depending on the particle size. More on this in [16].

Devices based on light scattering also allow direct particle counting, i.e., very small concentrations can be measured. However, as the scattering intensity decreases with



Fig. 15 Comparison of a standard NPTI-PN counter (PN-counter, AVL DiTEST GmbH, Austria) with the METAS reference device (NPET 3795, TSI Inc., USA). The NPET 3795 was traceably calibrated against the primary standard for particle number concentra-

d⁶ for particles smaller than the light wavelength, this does not work for the size range relevant here. The usual opacity measurement only provides useful results at high concentrations and is therefore out of the question. Both a measurement of the surface area (easily possible with DC devices) and the concentration of elemental carbon (with photoacoustic methods) would also be suitable metrics, as already mentioned above.

Since the various measurement methods react differently to different types of particles, it is important to calibrate the devices with suitable aerosols [48].

More information on the measurement methods can be found in [49, 50].

With the available experience after implementation of the PN-PTI procedure in 4 countries, after certification of 16 measuring instruments by NMI, METAS, and PTB and evaluation of several million measurements on LDV and HDV, the PN measurement at the PTI can be regarded as state of the art for the case of the diesel engine. There is a recommendation to adopt it in the Roadworthiness Directive of the EU Commission [51] and recommended by UNECE for worldwide application [52].

Further work concentrates on the measurement of gasoline engines, for which extensive experience is already available [53]. Figure 15 shows a comparison of a certified diesel PTI device with a METAS reference device. The agreement for this measurement is good. However, since the exhaust gas from gasoline engines contains considerably more volatile components, especially water, it cannot be concluded that devices developed for diesel emissions can also be used without additional measures for gasoline engines. Problems with nucleation occurred in other measurements. [54].

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tion. Left: comparison of the measured concentrations, right: counting efficiency of the PTI device as a function of concentration. This comparison has been done with CAST particles

6 Correlation of PN at Idling Speed to Type Approval Test Cycles

Strictly speaking, it is not necessary to correlate the measurement results of the PTI test with the emissions of vehicles in typical legal driving cycles. It is sufficient to show that the PTI test detects all significant technical faults influencing particle emissions with sufficient selectivity and that the subsequent repair of the damage has brought the vehicle back to the desired quality level or scrapped if repair proves not to be possible. The cost of this test must be so low that an annual 100% inspection of the fleet is justifiable.

Nevertheless, such a correlation is of course interesting and useful for the argumentation for the introduction of such a new procedure and provides a bridge for the cost/benefit analysis, which is always politically in demand.

These measurements are very laborious, but were carried out on a large scale at JRC and TNO and are shown in Figs. 16 and 17.

For the diesel (Fig. 16), there is a very good correlation for the cases without DPF; with DPF, the older measurements with pre-NPTI instruments show some scatter. Thanks to the measuring instruments now developed and certified in accordance with the NPTI standards of the NMI, however, these have shrunk considerably. This means that there is a good correlation between the emissions in the low idle mode and the WLTC measuring cycle, even three orders of magnitude below the current Euro 7 PN limit values. This diagram also shows that based on the official emission limit value for the type approval test in the WLTC cycle of 6×10^{11} km⁻¹, a pass/fail level of around 80,000 cm⁻³ should be selected. It also shows that many DPFs deliver better results by orders of



Fig. 16 Correlation of the PN-PTI measurement at idle speed of diesel engines to the legal emission measurement in the official test cycle [53]



Fig. 17 Correlation of the PN-PTI measurement at idle speed of gasoline engines to the legal emission measurement in the official type approval test cycle [14]

magnitude — an important indication for emissions policy to further reduce the limit value which is needed to avoid health risks.

For the gasoline engines (Fig. 17), the same test procedure produces a greater scattering. However, this result also clearly shows the need for a PN-PTI test. A striking number of the vehicles tested here exceed the official limit values. It is quite possible with this test procedure to reliably detect the highest emitters and thus massively improve the fleet quality. It is also clear that the restriction of Euro 6 legislation to DI engines was not sensible, PFI should also be included, and that the quality of GPFs has not yet reached that of DPFs, although intensive work is being done on this [55–58].

Further investigations have already shown that there are possibilities to improve the measurement method for petrol engines. For example, conditioning the engine at 60 °C,

increasing the idling speed to 2000 rpm, loading the engine at standstill by switching on the air conditioning and electrical consumers already lead to a significant reduction in scatter [58, 59]. The test method chosen by Mexico with a chassis dynamometer run also offers a very good solution, but requires a higher infrastructural effort [28].

7 Carrying Out the Vehicle Test

The test procedure chosen by the NPTI task force in the low idling speed of the stationary vehicle was thoroughly investigated by TNO and other partners of this group and finally laid down by NMI in a normative document [13] and thus adopted by the Netherlands and Belgium (Figs. 18 and 19). It allows a reliable measurement with two repetitions within 1 min, i.e., including setting up the measuring devices; no more than 3 min are required in the PTI process. The Swiss procedure is similar [60]; BRD has added two very useful elements to this: Checking the operating temperature of the engine — if this is below 60 °C, a short warm-up is carried out before the measurement and switching off the exhaust gas recirculation EGR.

Measurement can be carried out in different ways [61]. It is important that the effort involved remains low enough to justify an annual inspection of all vehicles in the fleet; experience in Switzerland has shown that this inspection of emission quality can be entrusted to certified workshops. This has the invaluable advantage that the workshops develop responsibility for the quality of emissions, that more specialist knowledge is generated, and that the know-how for repair the damage is generated.

8 Formation of Pass/Fail Criteria

If the diesel engine is adjusted and maintained in accordance with the regulations and is operated with the prescribed fuel and low-ash lubricating oil and is equipped with a particle filter fulfilling VERT [62, 63] or similar requirements, the number concentration of solid particles greater than 23 nm should not exceed a value of 10,000 cm⁻³ at low idle, usually even a level of < 1000 cm⁻³ is maintained. However, in order to form a pass/fail criterion suitable for practical use, the scattering of the measuring method, the measuring conditions and the measuring devices must also be taken into account. This results in a value of 50,000 cm⁻³, and this value is proposed by VERT in the Technical Guideline TA 24 [11].

Figure 20 shows how the choice of the pass/fail criterion affects the failure rate and thus the costs of quality assurance for the Swiss construction machinery fleet.

It is also clear that more DPFs have failed in the old retrofit fleet (from 2000) than in the construction machinery Engine speed

Fig. 18 Proposal for the measurement procedure as it was then adopted in the NPTI standardization [13]. The stationary vehicle is operated in low speed, a procedure that is also suitable for road side checks by the police



0.0E+00

from AVL DiTEST or at a standstill on the roadside during a police check

brought onto the Swiss market by OEMs from around 2012. Under EU law, DPFs are not mandatory until Stage V from 2019.

The pass/fail value in high idling specified in this diagram for Swiss construction machinery was set at 250,000 cm⁻³ [18] and is derived from the type test value $1 \times 10^{12} \text{ kWh}^{-1}$.

The correlation to the type test for passenger cars according to Fig. 16, on the other hand, leads to a pass/fail for PTI at low idle of approx. 80,000 cm⁻³ for diesel engines. For gasoline engines, the correlation shows significant scattering (Fig. 17). The situation there is more complicated, as measurements at low idle, idle at elevated speed, and additional load show significant differences [64]. Nevertheless, a limit in the same range makes sense.

The current rejection values of 10^6 cm^{-3} in the low idle range set for PTI in various countries are politically motivated in order to initially capture the worst emitters when the procedure is introduced and will certainly then be reduced to 250,000 cm⁻³ and perhaps even to 100,000 cm⁻³, a value that is already used for first pass in Switzerland today.

For petrol engines, the pass/fail criterion has still to be elaborated, but based on our actual knowledge, it should not be much different from diesel.

Fig. 20 Failure rates of the Swiss construction machinery fleet as a function of the pass/fail criterion [21]

6.0E+05

8 0E+05

4.0E+05

PN [#/cm3]

9 General Validity for the Quality Assessment

2.0E+05

This paper aims to make it clear that the emission quality control of combustion engines is best achieved by measuring the particle number concentration; it does not only record the emission of solid carbon particles, but also that of the PAHs deposited there and the highly toxic heavy metals that also contribute to the risk. Therefore, this is no longer just about monitoring the functionality of DPF and GPF particle filters; it goes much further, namely to the many sources of particle formation. This includes not only combustion with its many influencing factors, such as injection, supercharging, and exhaust gas recirculation, but also wear, lubrication deficits, unsuitable maintenance, use of unsuitable fuels, incorrect control and regulation settings, hardware and software errors in exhaust gas aftertreatment, inadequate maintenance, and, unfortunately, deliberate manipulation of hardware and software. It seems that the PN emission is of

1.0E+06

central importance for many different deterioration effects of the combustion engine and that this highly sensitive signal can be used to detect many failures influencing toxic emissions. It therefore concerns all fleets of combustion engines, diesel and gasoline, with and without exhaust aftertreatment, on-road and off-road, light duty, heavy duty, stationary, and NRMM.

Experience to date suggests that the failures generally accumulate in a small number of vehicles and can reach high PN emissions in the superposition, the so-called super polluters, whereby the causes can be quite different. However, they are thus reliably detected and can be quickly eradicated by repair, replacement of filter retrofit to the extent desired in terms of emissions policy.

It should be noted that this not only makes a major contribution to reducing local health risks, but also makes a significant and important contribution to reducing global warming [65, 66].

10 Alternatives to PN-PTI

In addition to roadside checks by the police, which are defined by law but are limited to a few individual cases, two additional methods are also discussed used, namely "chasing" and "remote sensing."

Chasing means exhaust gas measurement by a chasing vehicle and has recently been perfected in theory and practice [66, 67]. Today, it can be used not only on special routes for research purposes, but also for police checks, e.g., on highways. There is no doubt that it can be used to identify high-emission vehicles. The procedure has proven its worth in order to substantiate facts in the case of a preliminary suspicion, to be able to take the vehicles out of circulation and to investigate manipulative interventions more closely [68]. However, its use remains limited to individual cases, and if a penalty is to be imposed for excessive emissions, the vehicle will have to be referred to a periodic emissions test, as this is the only place where the measurement procedure and pass-fail limit value are clearly regulated by law.

Remote sensing typically refers to the emission monitoring of vehicles in road traffic using a laser beam directed across the road or a vertically aligned beam in bridges or tunnels. Concentrations of gaseous emissions can be determined by NDIR methods and particles > 400 nm by scattered light methods [69, 70]. Also, BC measurement by photoacoustics has been demonstrated and even PN counting [71]. As a large number of vehicles can be recorded, the method is very well suited to identifying trends in emissions depending on the years of enrolment or even the manufacturers and vehicle types [72]. On the other hand, although it is possible to detect high emitters, it will not be possible to determine the degree of exceeding the emissions limits with the legally required certainty, as the operating status of the vehicle is not known and the overlapping of emissions from other vehicles cannot be excluded. Consequently, here too, the matter is referred to the PTI where the final pass/fail decision must be taken.

11 Recommended Procedure for Use with All Vehicle Fleets

Many countries already have a good PTI system in place today, in which not only safety elements of the vehicles are checked, but also pollutant emissions. The EU's decision in 2012 to delegate this control entirely to the on-board diagnostics (OBD) system and thus dispense with independent controls was fortunately not adopted outside the EU and has now been corrected [51].

Upgrading the PTI process by a PN measurement to get the dirty tail information is very simple. For gasoline engines, it may be useful to test at high idle speed and with some electric load. This prolongs the PTI process time by a maximum of 3 min and high emitters will be identified right away. Of course, this simple test does not show directly what the problem is. But if the vehicles are then tested in a workshop, the problem can be identified and corrected.

Abbreviations AeroSolfd: EU-project to minimize particle emissions from petrol engine; AFHB: Abgasprüfstelle und Motorenlabor der Fachhochsschule Bern; AVL: Anstalt Verbrennungsmotoren List, Graz; BAFU : Swiss Federal Office for the Environment; BRD: Federal Republic of Germany; CALAC: Clean Air for Latin America's Cities; CE: Counting efficiency; CH: Confoederatio Helvetica = Switzerland; CPC: Condensation particle counter; CO: Carbon monoxide; CO2: Carbon dioxide; COP: Conformity of production; DC: Diffusion charging sensor; DI: Direct injection; DiSCmini: Portable PN measuring system; DOC: Diesel oxidation catalyst; DPF: Diesel particle filter; EGR: Exhaust gas recirculation; ETH: Eidgenössische Technische Hochschule (Zürich); GPF: Gasoline particle filter; HC: Hydrocarbons; HDV: Heavy duty vehicle; ICE: Internal combustion engines; HORI-ZON: EU research framework; ISC: In-service conformity; JRC: Joint Research Center of the EU Commission, Ispra, Italy; LDV: Light duty vehicle; LRV: Swiss Ordinance on Air Pollution Control (LRV); METAS: Swiss Federal Institute of Metrology; MIL : Malfunction indicator light - part of OBD; NanoMet: PN measuring system consisting of PAS &DC; nm: Nanometer = 10 m^{-9} ; NDIR: Non-dispersive infrared analysis; NMI: Netherland Metrology Institute; NOx: Nitrogen oxide NO+NO₂; NPTI: VERT guided task force for (new) periodic technical inspection on PN-basis; NPET : First NPTI-certified PN measuring device ; NRMM: Nonroad mobile machines (e.g., construction); OEM: Original equipment manufacturer; OBD: On-board diagnosis; PAH: Polycyclic aromatic hydrocarbons; PAS: Photoelectric aerosol sensor; P/cm³: Particles per cubic centimeter; PFI: Port fuel injection; PM: Particulate mass; PM2.5: Particulate matter (mass) below 2.5 µm; PMP: Particle measurement program of the UNECE; PN: Particle number, particle count; PN-PTI: Periodic technical inspection based on the number of particles; PTB: Physical-Technical Federal Institute of Germany; PTI: Periodic technical inspection; R²: Error square; RDE: Real driving emission; RH: Relative humidity; SCR: Selective catalytic reaction/reactor; SEDEMA: Secretaria del Medio Ambiente; SERI: Swiss State Secretariat for Education, Research, and Innovation;

STVZO: Road Traffic Licensing Regulations of the Federal Republic of Germany; TCS SE : Touring Club Schweiz, Sektion Biel/Bienne-Seeland; TNO: Organization for Applied Science of the Netherlands. www.tno.nl; TÜV: Technischer Überwachungsverein (Germany); TWC: Threeway catalyst; UNECE: United Nations Economic Commission for Europe; VAMV: Swiss ordinance on exhaust gas analyzers; VERT: Verification of Emission Reduction Technology; Association; www.vert-certification.eu; VPR: Volatile particle remover

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Data Availability Date for this study are to be found in the VERT archives and can be shared with interested parties.

Declarations

Ethical Approval Not applicable since we are only dealing with purely technical matters.

Competing Interests The authors declare no competing interests.

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