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Industry comments on proposed particulate measurement techniques

OICA contribution to PMP

Part 1: Synthesis Report

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Summary

Data and information on several techniques for measuring vehicle PM emission are presented in this report.

This review shows that only the gravimetric method fulfils the measurements requirements for a technique to be applied for type approval. Other mass based methods may have the potential to fulfil these requirements.

The investigated new measurement instruments can, as yet, only be used for the qualitative assessment of particulate number and size distribution as a relative comparison. They are still far from achieving the absolute quantitative measurement of these parameters. The measured particle number and size distribution can be manipulated with a careful choice of test conditions. Any new particulate measurement methodology should fulfil the same quality criteria as the current gravimetric method. Based on the above assessment, gravimetric measurement is considered to have the highest potential for future development.

1. Introduction

At the 41st meeting of ECE-GRPE (16. – 19. 01.2001, Geneva) it was agreed to set up an ECE-GRPE ad hoc WG PMP to develop a new particulate assessment and measuring system for application during exhaust emission type approval testing of light-duty vehicles and heavy-duty engines.

The European automotive industry is actively involved in the PMP program and is doing intensive research work in this respect. It has been suggested that a new particulate measurement methodology may be introduced in a future exhaust emission regulation with a set of limits in parallel to the existing particulate measurement legislation based on the principle of gravimetry (determination of the mass of particulate matter (PM)).

The first investigations (about 20 years ago) into a possible health risk of diesel particulates were carried out with toxicological studies with the support of the automotive industry. The results[1] show that animal experiments cannot be applied to human beings. Comparative studies between monkeys and rats prove furthermore that the deposition of particulates in lungs differs considerably. In the lungs of monkeys, that are very similar to those of humans, particulates are mainly deposited in the tissue and cell interspaces without causing inflammations that often precede the formation of tumours. In rat lungs, however, the particulates are mainly deposited in the lung pulmonary alveoli causing changes in the tissue and inflammations. Therefore within the international discussion the animal experiment results are not considered valid for application to humans. Hence they are unsuitable as a basis for risk assessment for humans.

The results of the subsequent epidemiological studies are not clear enough to make a conclusive statement[2]. Aside from a few exceptions the studies are based on measurements of the exposure in relation to the total particulate mass. This requires further clarification before new legislation can be introduced.

The independent HEI[3] states[4] that the results of an extensive study "indicate that epidemiological evidence of PM's effects on morbidity and mortality persist even when the alternative explanations have been largely addressed".
Although HEI places special emphasis on the necessity of
?? further research to clarify the impacts of the health effects and
?? caution in the use of these results in the political field
the above statement is often used in isolation and an immediate drastic reduction of particulates is demanded as a precaution.

Furthermore, the HEI[5] commented that both the ultrafine (0.010-0.100 µm) and fine (0.010-2.5 µm) particle fractions have shown associations with human mortality; however 'no clear pattern of association indicates relative or temporal differences between ultrafine and fine particles'. This essentially means that there is currently no clear evidence to preferentially address air quality or emission standards for ultrafine particles, as compared to the existing standards for fine PM.

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[1] M. Spallek: Stellenwert der Tierversuche zu Dieselmotoremissionen aus arbeits- und umweltmedizinischer Sicht, ErgoMed 4 (1997), 102-104

[2] M. Spallek: The effects of exhaust emissions on human beings – A never ending story?, VDA Technischer Kongreß, 20./21.09.1999, Frankfurt, S. 201-209

[3] HEI = Health Effects Institute, Cambridge MA, USA (www.healtheffects.org)

[4] Airborne Particulates and Health: HEI Epidemiological Evidence, HEI Perspectives, Cambridge MA, USA, June 2001

[5] HEI Research Report 98

The US- EPA[6] currently does not perceive a need for a new metric for PM emission regulation, other than PM mass, due to the lack of health effects evidence. This statement is based on the acknowledged health effects related to PM mass, which has led to implementation of ambient PM mass standards and emission inventories. However, the US-EPA remains open to considering alternative methods of determining the PM mass emissions[7].

In principle, the automotive industry is of the opinion that the reduction of particulate matter which has already been achieved and which may still be possible in future can be already considered as a health precautionary measure.

Following the outcome of the research activities of BMW, Bosch, DaimlerChrysler, Ford, JAMA, Opel, Renault, Volkswagen and ADA[8] concerning the evaluation of the different instruments is outlined and discussed below:

2. Results of measurement programmes from ADA, BMW, Bosch, DaimlerChrysler, Ford, JAMA, Opel, Renault, and Volkswagen

2.1 Influences of engine, fuel and measurement and sampling conditions on aerosol measurements

Measuring aerosols in terms of size, number and surface is much more complex than measuring mass, since aerosol measurements determine integrated properties which are based on individual particles distributed in a carrier gas. Number concentration and morphology change continuously due to external influences such as temperature, dilution and gas velocity. Therefore aerosol measurement (and this includes measurement of engine exhaust) is just a momentary photograph at time and location of measurement with an unknown relationship with the real world situation.

?? Nucleation (fuel sulphur) and artefact particle effects

NSD[9] emission measurements results from Ford, BMW, Volkswagen, Opel and ADA show that nucleation particles occur in the <10 nm to 30 nm size range. The nucleation mechanism and chemical composition of these particle droplets is highly complex and not well understood. Nucleation particles occur during cooling and dilution. However, currently, there is no method available to reproduce the atmospheric dilution conditions in the laboratory. The formation of nucleation particles is dependent on the fuel sulphur content (see Figure 1 and 2), engine load and catalyst activity (Figure 3 and 4) and will be reduced by the introduction of future low sulphur fuels.

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[6] EPA Report: Health Assessment Document for Diesel Engine Exhaust

[7] CRC workshop San Diego, EPA presentation M. Spears

[8] ADA Abgaszentrum der Automobilindustrie

[9] NSD = Number Size Distribution

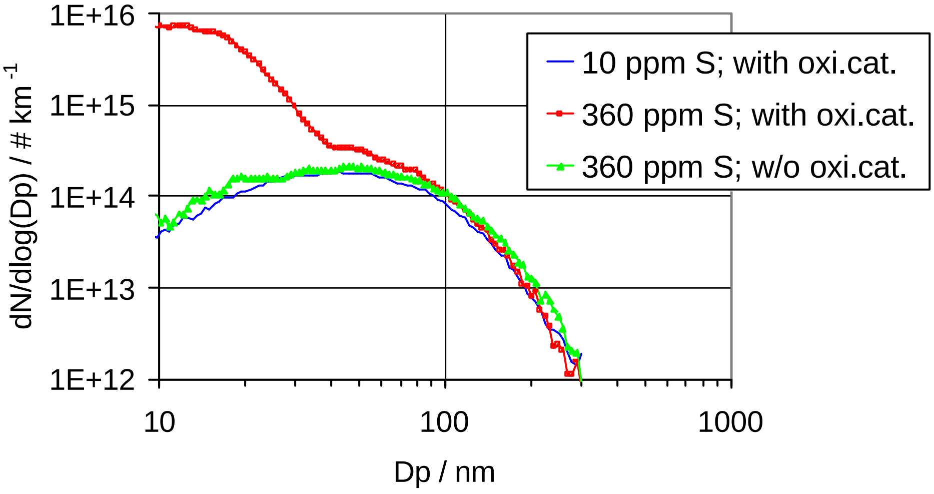

Figure 1: Real-world particle size distributions measured by SMPS[10] at 120 km/h chasing the exhaust plume of a diesel test vehicle operated with 360 ppm S fuel with and without oxidation catalyst with a mobile laboratory (red squares and green triangles). The blue solid line represents a measurement with 10 ppm S fuel with oxidation catalyst, showing the absence of nucleation particles. The distance between test vehicle tailpipe and sampling inlet was 14 m (0.4 s). Ambient temperature was 20 °C, relative humidity 60 %. Nucleation particles occurred only if an oxidation catalyst and high sulphur fuel were applied. *(Vogt and Scheer, 5th Int. Workshop on Nanoparticle Measurements, Zürich, 6.-8.8.2001, Proceedings Bundesamt für Umwelt, Wald und Landschaft, Bern (2001)).* (Source: See Annex 5.5, Ford)

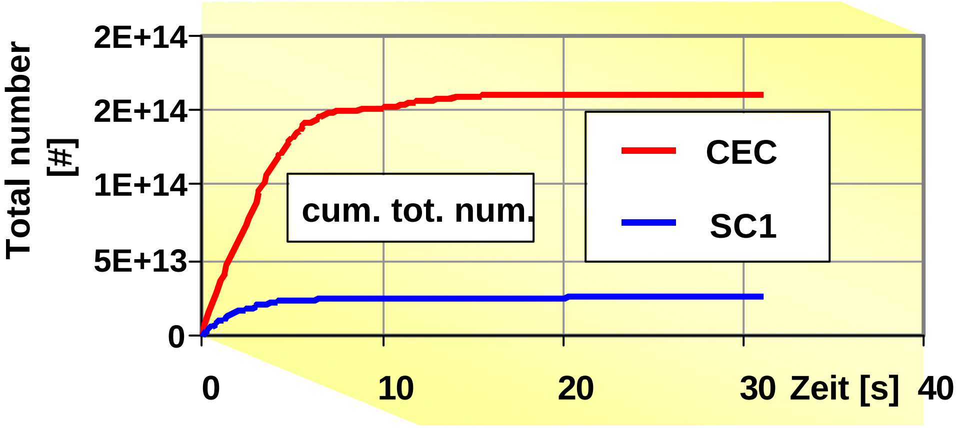

Figure 2: Braking 120 to 0 km/h with down shifting from 5th to 1st gear within 19 sec, OPEL Astra with 2.0 DTI diesel engine. The figure shows the accumulated number of particles (TDMPS[11]) during braking and subsequent idling for a CEC fuel (CEC) and the Sweden Class 1 fuel (SC1) . Despite the fuel cut off under braking conditions, the number of particles increases. This is attributed to residual fuel and deposits and is a much smaller effect with the low sulfur Sweden Class 1 fuel. (Source: Opel)

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[10] SMPS = Scanning Mobility Particle Sizer

[11] TDMPS = Transient Differential Mobility Particle Spectrometer

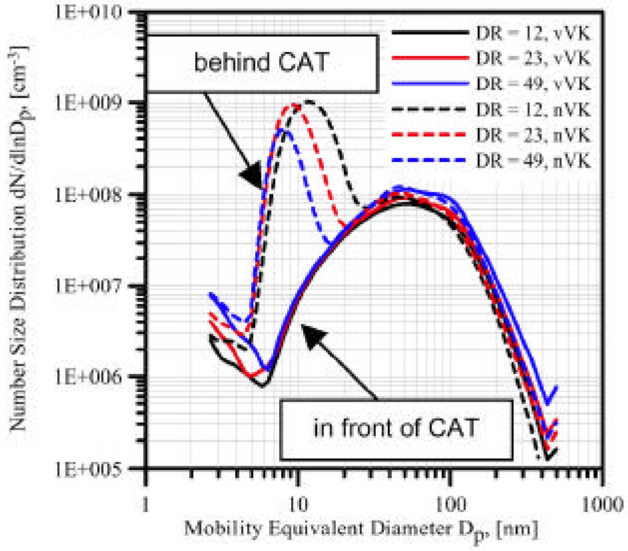

Figure 3: Two sampling points in front of the catalyst (vVK) and after the catalyst (nVK) are shown. In the range above 30 nm, the shapes of the curves of sampling in front and after the catalyst are nearly the same. Below 30 nm, the strong nucleation effect is clearly influenced by dilution ratio (DR) effects.
The particle size distributions were measured with a twin SMPS with a Faraday-Cup Electrometer. The tests had been carried out on an engine test bed with a BMW Diesel engine at an operation point corresponding to a vehicle speed of 120 km/h with 210 ppm sulphur fuel content. (Source: See Annex 5.2, BMW)

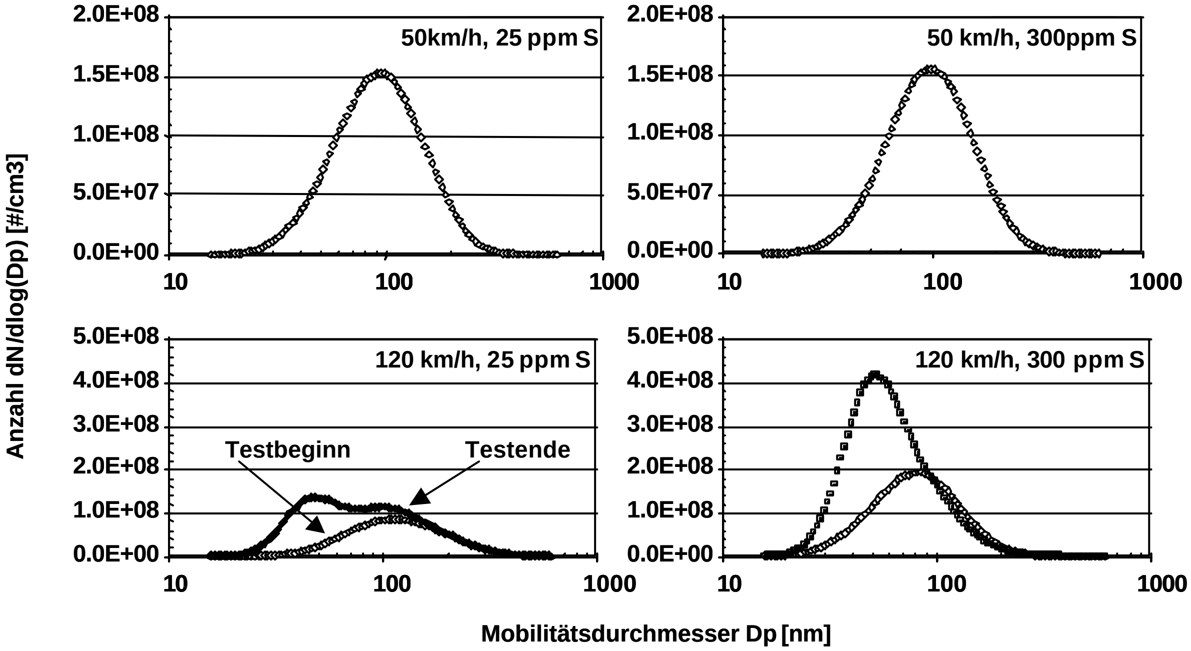

Figure 4: Comparison of SMPS Diesel particulate size distribution at 50km/h (upper diagrams) and 120km/h (lower diagrams) using diesel fuels containing 25 ppm sulphur (left diagrams) and 300ppm sulphur (right diagrams). At 50 km/ h the size distribution is monomodal with a maximum particulate size of approx. 90 nm. No significant influence of the sulphur content of the fuel can be observed. The results at 120 km/h show a clear difference when using fuels with different sulphur contents. With high sulphur content a condensation particulate mode can be observed which finally dominates the size distribution. (Source: See Annex 5.6, VW)

Large artefact effects showing high numbers of nucleation particles have been documented (see Figure 5).. The "false" measurements (result influenced by high number of artificial particles) occur due to pyrolysis of tube connectors, or due to the evaporation and condensation of deposit material from the transfer line.

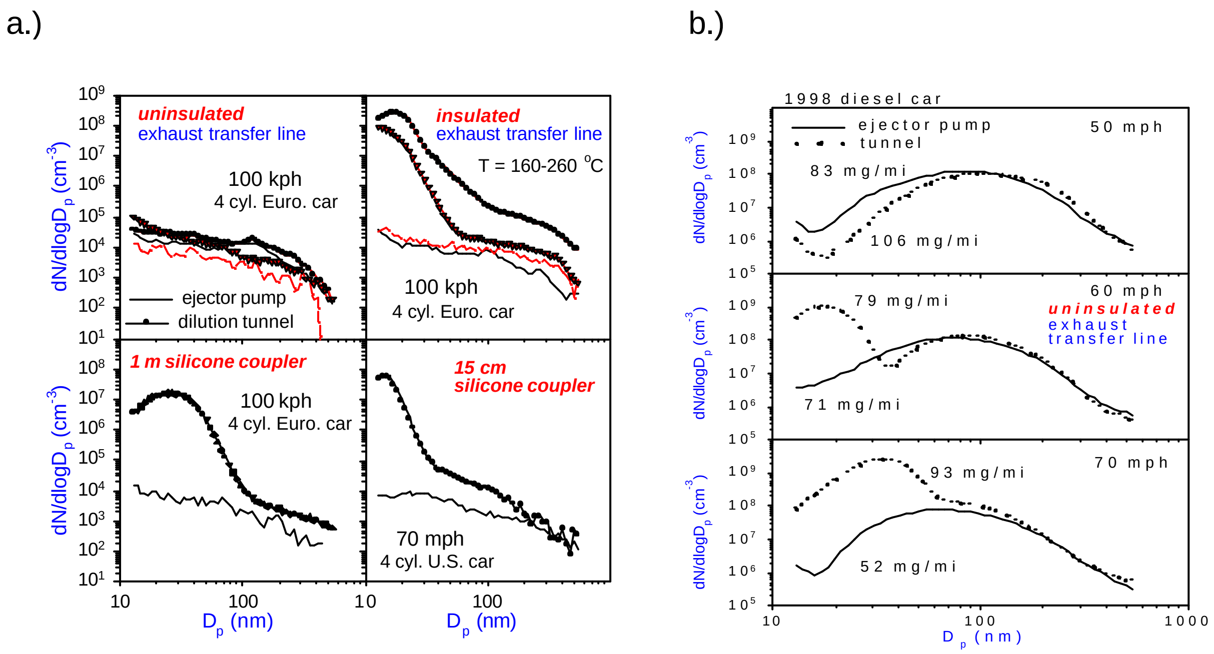

Figure 5: Comparison of particle size distributions (SMPS) for a European gasoline car a.) and a 1998 diesel car b.) taken by ejector pump versus dilution tunnel when using a 1) *not insulated* transfer line, 2) *insulated* transfer line, 3) *not insulated* transfer line plus a 1 meter silicone rubber coupler, and 4) *not insulated* transfer line plus a 15 cm silicone rubber coupler. It is shown that with excessive heat, high numbers of artificial particles occur in the dilution tunnel which are absent when sampled directly from the tailpipe *(Maricq et al., SAE 1999-01-1461)*. All concentrations are converted to tailpipe concentrations. The tunnel background level was in the order of 5000 particles cm-3 and was subtracted. (Source: See Annex 5.5, Ford)

A new PM measurement metric which is sensitive to nucleation particles is potentially susceptible to large measurement artefacts and should therefore not be considered further.

?? Use of thermodenuder TD

The thermodenuder is a device to remove volatile particles and is being considered to overcome measurement errors caused by the occurrence of nucleation particles. In some cases this might be an optimistic assumption because

- the efficiency for removal of volatile particles is not 100 %;
- size dependent losses of non-volatile particles occur.

A thermodenuder has a particle size-dependent transmission efficiency which is also related to the TD carbon bed temperature. This means that it is not at all suitable for use in combination with size-unresolved total particle count (see Figures 6 and 7).

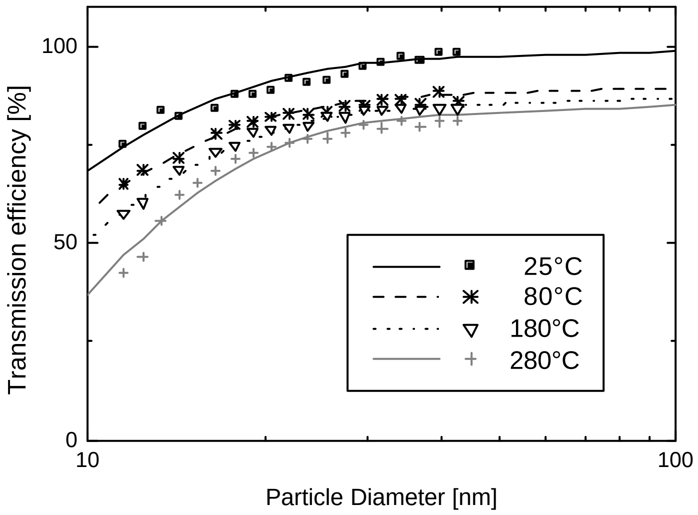

Figure 6: Particle size-dependent transmission efficiency of silver particles within a thermodenuder for different temperatures, SMPS measurements (Wehner et al. J. Aerosol Sci. 33, 1087 (2002).

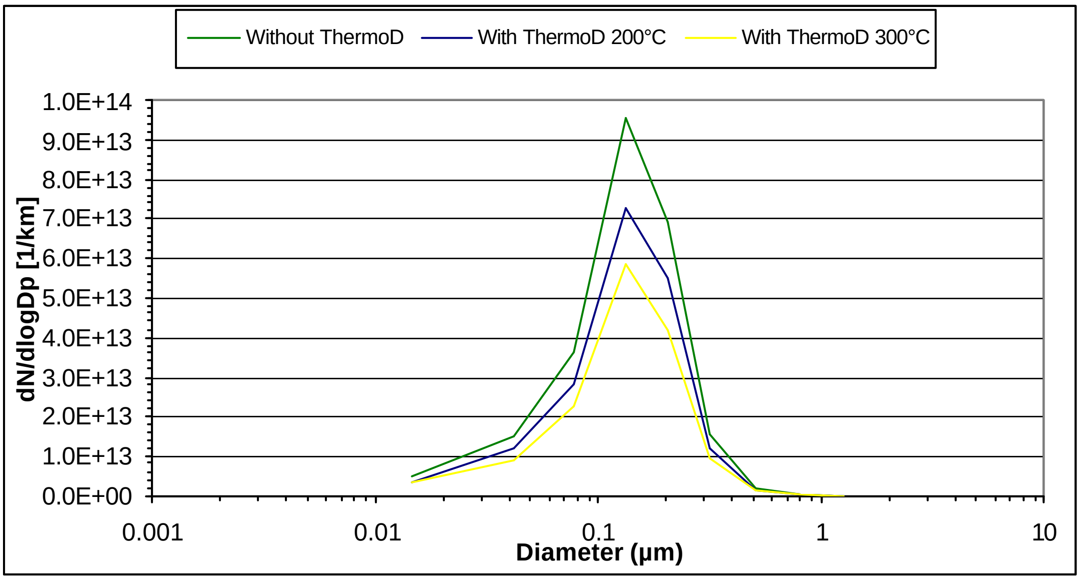

Figure 7: Relative effects of a thermodenuder and different temperature conditions on the size-number distribution (ELPI[12] - greased sintered plates - sample from the CVS - secondary dilution (x 10) with N_2) of Diesel exhaust particles as measured over an hot NEDC test cycle (Euro 3 - Common Rail Turbo-charged Diesel car - Fuel : 270 ppm S). The total particle numbers are drastically affected by the presence of the thermodenuder and the carbon bed temperature conditions (5 * 10^{13} /km without heating ; 4 * 10^{13} /km with (200 °C) and 3 *10^{13} /km with (300 °C) respectively) . In addition, the trapping efficiency is strongly size dependent and will be probably influenced by maintenance and ageing. (Source: (Source: See Annex 5.7, Renault)).

The efficiency of a thermodenuder to remove volatile particles and the size-dependent transmission efficiency for solid particles is dependent on various parameters such as flow rate, inlet and carbon bed temperature, chemical composition of the aerosols (see Figure 8), residence time in heating and adsorption section, carbon bed absorption

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[12] ELPI = Electrical Low Pressure Impactor

capacity, design etc. In principal it is necessary to validate the performance of a thermodenuder both before and after every test as there is no obvious means of determining when the carbon in the adsorption bed is saturated.

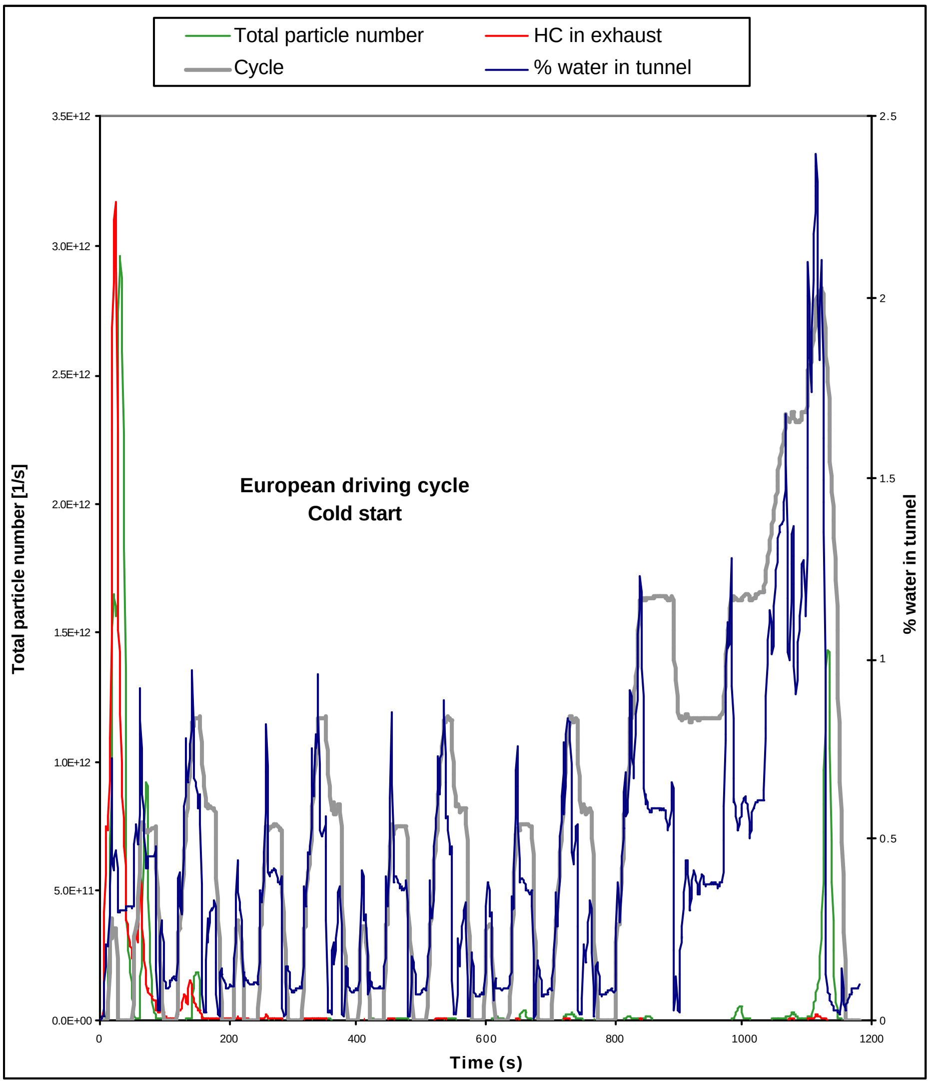

Figure 8: Total particle number (3.3x10^{11} /km ELPI - greased Al plates - sample from the CVS - secondary dilution (X 10) by N2) of a typical Gasoline exhaust as measured (without a thermodenuder) over a cold NEDC test cycle (Euro 3 - MPI Gasoline car ; Fuel : 130 ppm S).

A thermodenuder (250 °C) used under the same test conditions will strongly reduce the total particle number (1.4x10^{11}/km) mainly as a result of trapping condensed water aerosols: Under hot NEDC conditions (with extremely low residual HC emissions – i.e. efficient after-treatment), the only significant effect of the thermodenuder is on the water based particle emissions. This occurs towards the end of the NEDC cycle. It demonstrates that the performance of a thermodenuder is directly influenced by the test conditions and will probably reveal an unreliable behaviour over extended periods of time, depending on the maintenance scheme. (Source: See Annex 5.7, Renault)

A combination of thermodenuder with size-unresolved total particle count cannot give valid data, because of size-dependent particle losses, and likely chemical species dependency on denuder efficiency.

Because of the many parameters that must be kept constant and verified by regular calibration, a thermodenuder is in principle not suitable for regulatory measurements.

?? **Sampling location and residence time (coagulation) effects**

The measured number/size distributions may also be dependent on the sampling system, even in the absence of nucleation particles. Depending on sampling location and the residence time, particles may coagulate and shift the size distribution to somewhat larger diameters, whilst the total particle number is significantly reduced (see Figures 9, 10, 11, 12).

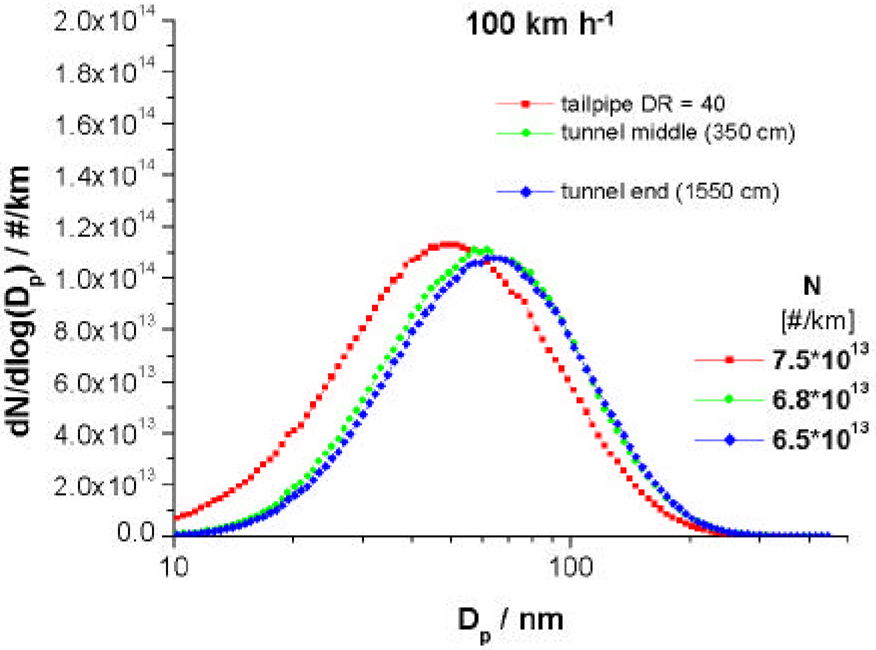

Figure 9: Comparison of particle size distribution (SMPS) measured at three sampling locations with a rotating disk diluter. The diesel vehicle was running at 100 km h^{-1} constant speed. The diluter was set to a dilution ratio of 1:40. At the tunnel the total dilution ratio was 1:440. Under these conditions the measured particle size distribution shifted to larger diameters and the total number decreased with longer residence times. *(Vogt and Scheer, 5th Int. Workshop on Nanoparticle Measurements, Zürich, 6.-8.8.2001, Proceedings Bundesamt für Umwelt, Wald und Landschaft, Bern (2001)).* (Source: See Annex 5.5, Ford)

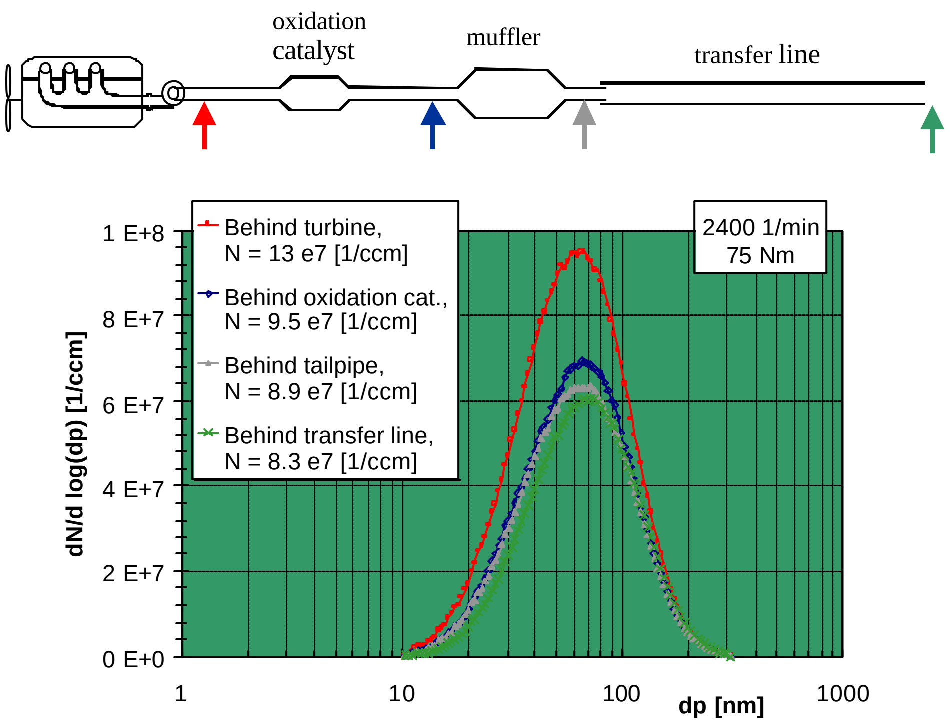

Figure 10: Size distribution and count median diameter, measured at various sampling locations with a DMPS[13] in the heated path (350 °C) and a one stage ejector dilution system. The decrease in number concentration and the slight shift in particle size is caused by agglomeration, diffusion and thermophoresis. The engine was a 3 l, 6 cylinder, common-rail direct injection diesel engine from a passenger car, operated with low sulphur fuel. (Source: See Annex 5.3, Bosch)

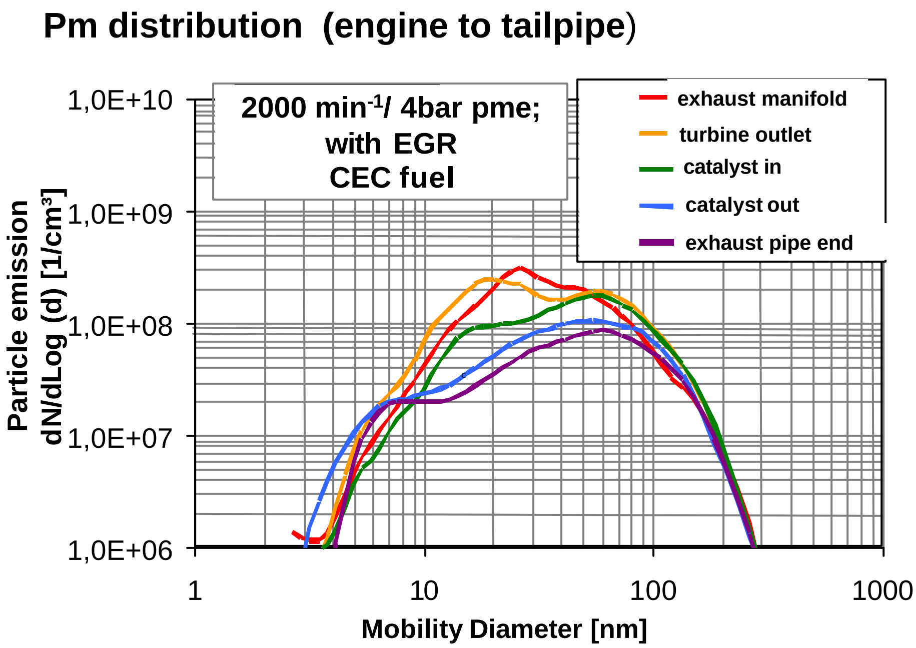

Figure 11: Size distribution and count median diameter measured along the exhaust pipe system of the Opel X20 DTdiesel engine. The size distribution is affected by the coagulation process which occurs along the exhaust system. In the catalyst formation of nucleation particles through sulphur oxidation occurs. Measurement was done with a 10 channel Transient Differential Mobility Particle Spectrometer (TDMPS) in collaboration with the University of Wien. (Source: Opel)

There is a different aerosol residence time for the undiluted gas which is dependent on the length of the transfer lines. Interaction of the aerosol with the surface of the lines and agglomeration increases with longer lines (see Figure 12).

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[13] DMPS = Differential Mobility Particle Sizer

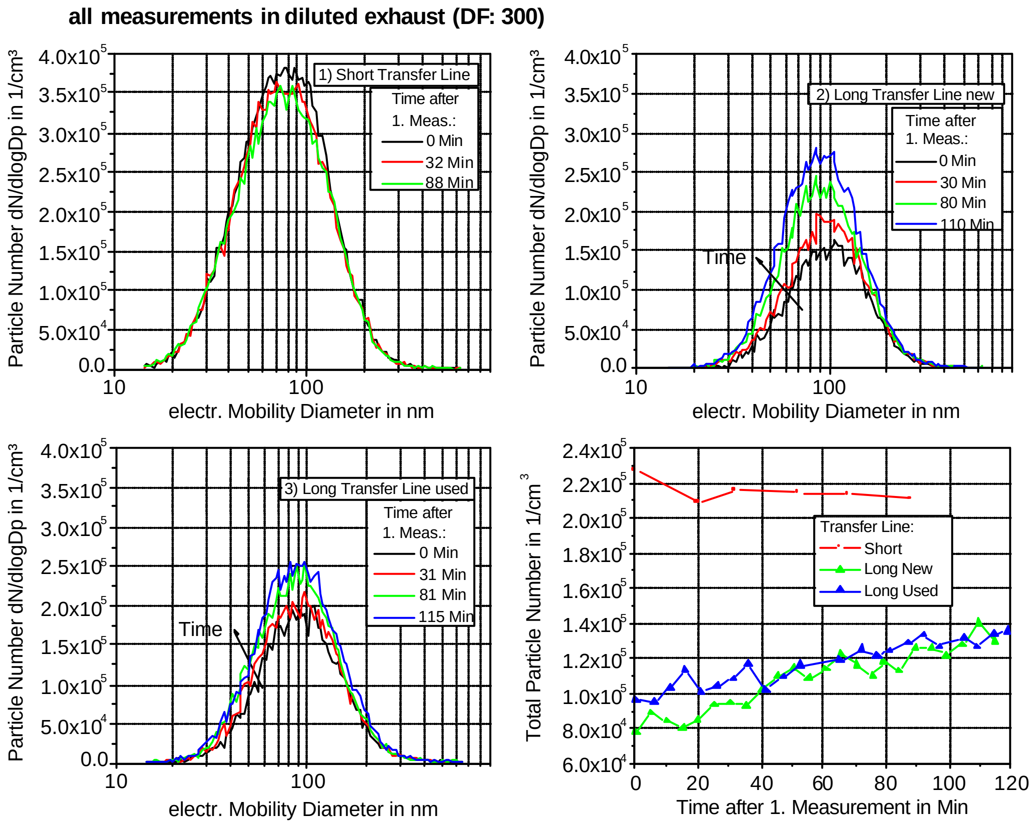


Figure 12: Number size distribution measured with SMPS combined with dilution system NanoMet rotating disc. Use of two transfer lines (Material: stainless steel) with different lengths (Short: 0.3 m , long: 1.7 m). The long line was tested in two different conditions: new (clean) and after a few days of being used for measurement (contaminated with diesel exhaust). The heated transfer line (120 °C) transports the undiluted gas from sampling position to the diluter. The engine operating point is held constant during all measurements. With the short transfer line, the NSD and particle number concentration remain nearly constant over measuring time. In comparison, the long line (1.7m) shows NSD changes and number concentration increasing with time, although the concentrations measured with the short line are not reached even with an extended measurement time. Changes of NSD over time with a new line are higher than with a used line. (Source: See Annex 5.1, ADA)

A measurement metric based on 'solid' particle number is problematic because of the influence of aerosol dynamics on the measured value.

?? Particle morphology influences on size measurements

TEM (Transmission Electron Microscopy) measurements (see Figure 13) suggest that morphology changes of particles induced by changes of operating condition and fuel quality can lead to results which are not reproducible. The method of classifying particles, i.e. the aerodynamic diameter (i.e. ELPI) and the electrical mobility diameter (i.e. SMPS) have a large influence on the measured particulate number and size distribution (see Figure 14).

Various measurement technologies are based on different physical quantities. During measurements the idealised physical behaviour of the particles (e.g. constant density, spherical shape, ...) cannot be assumed. Therefore, the results depend on time, location, operation point and fuel quality.

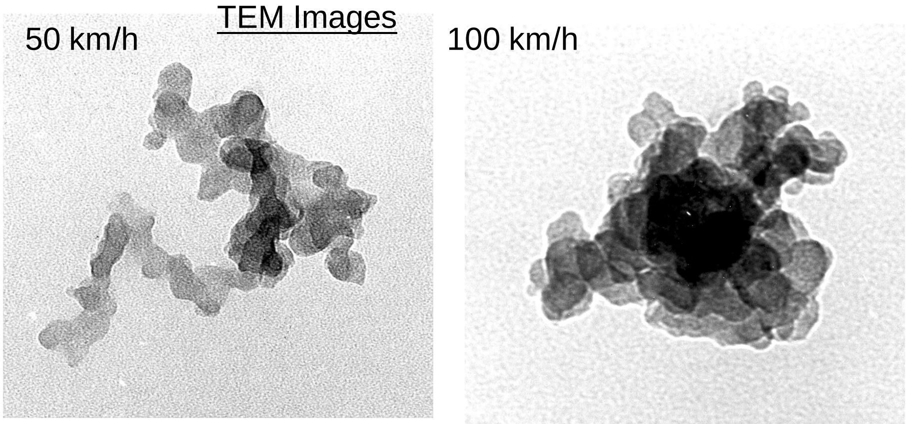


Figure 13: TEM images of Diesel particles samples at 50 and 100 km/h load. (Source: See Annex 5.6; VW)

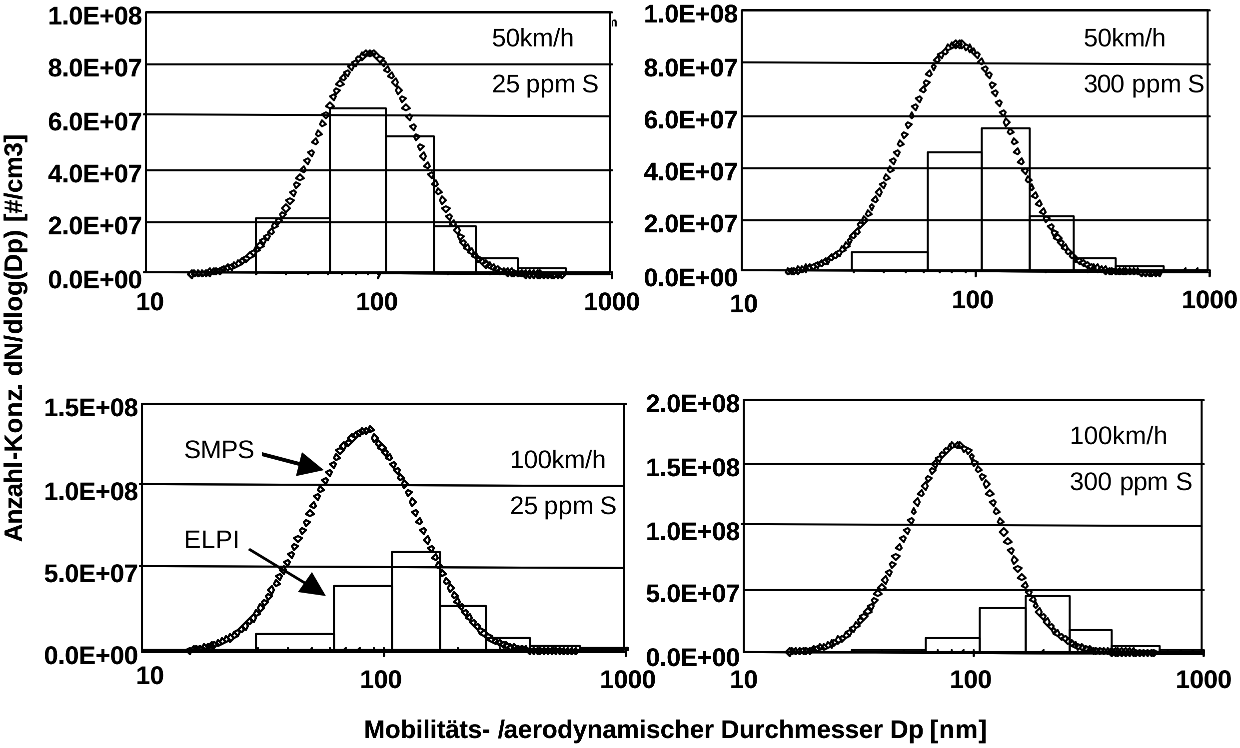


Figure 14: Comparison of parallel SMPS and ELPI measurements under different loads and fuel sulphur content. With increasing load and sulphur content the ELPI shows a decrease of the particulate number and a shift of the distribution maximum towards bigger particulate diameters. Whereas in the SMPS the distribution maximum remains constant and only the number increases. Load and fuel sulphur induced morphology changes are thought to be the reasons for this effect. (Source: See Annex 5.6, VW)

Engine load and fuel dependent particle morphology changes can lead to unpredictable aerosol measurement effects. Examples include aerodynamic and mobility aerosol behaviour. Metrics influenced by this type of effect are neither reliable nor repeatable.

?? Dependence of particle size distribution on injection pressure

The influence of the injection pressure on the particle size distribution is being discussed. The number size distribution in the exhaust of a high pressure, direct injection diesel engine has been measured over a wide range of injection pressures (see Figure 16) When the original injection pressure of 550 bar is varied over a range of 250...1600 bar and the engine torque, the soot and the NOx level are kept stable, no effect of injection pressure variation on particle size distribution can be found.

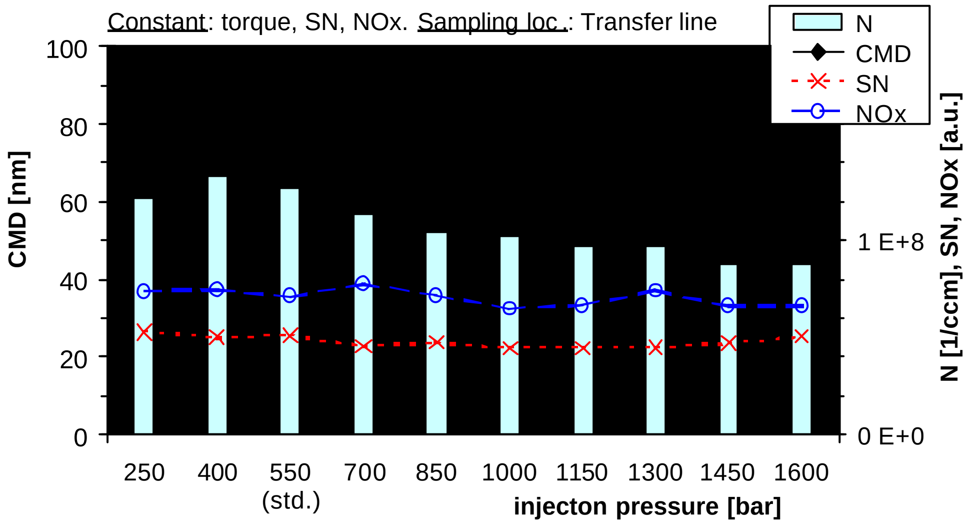


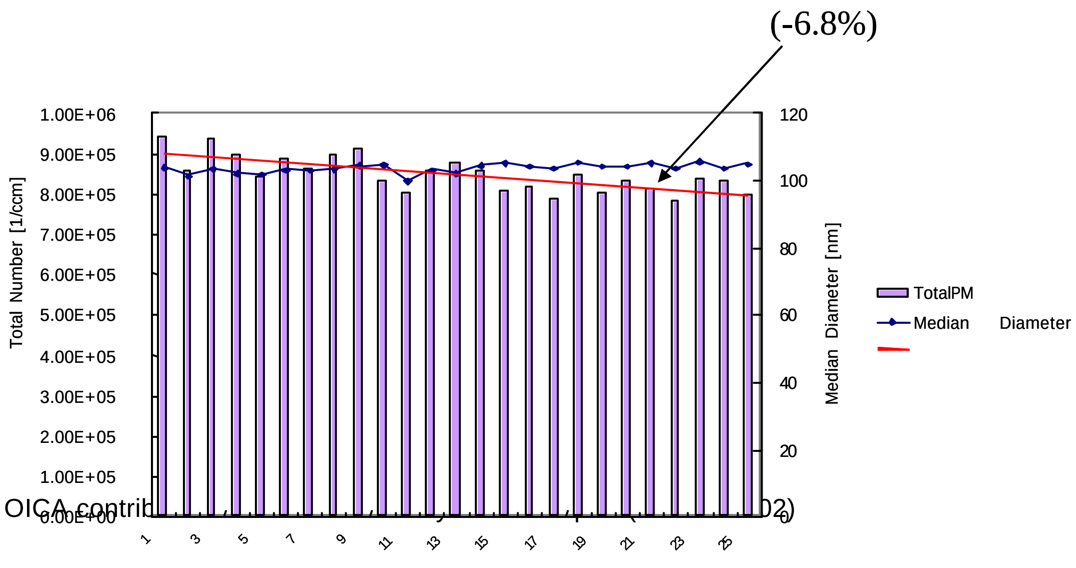
Figure 16: Particle size (CMD) and number concentration (N) versus injection pressure; n = 2000 1/min, M = 82 Nm (--> v = 100 km/h). Torque, NOx and the smoke number were set to their original value, after changing the rail pressure (standard values at this engine operation point: rail pressure = 550 bar, smoke number (SN) = 1.0, NOx = 140 ppm). Sampling location was behind a transfer line. The number size distribution has been measured with a SMPS and a one stage ejector dilution unit by Bosch. The measurements were carried out on a stationary test bench with a 3 l, 6 cyl., common rail direct injection engine from a passenger car. (Source: See Annex 5.3, Bosch)

With a test procedure of practical relevance, no correlation between particle size and injection pressure of a modern, direct injection diesel engine was identified. As particles are formed in the gas phase, their size is not related to the droplet size in the diesel spray or to the injection pressure. Size resolved particle measurement gives no additional information and can therefore be replaced by mass measurements.

2.2 Evaluation of instrumentation systems

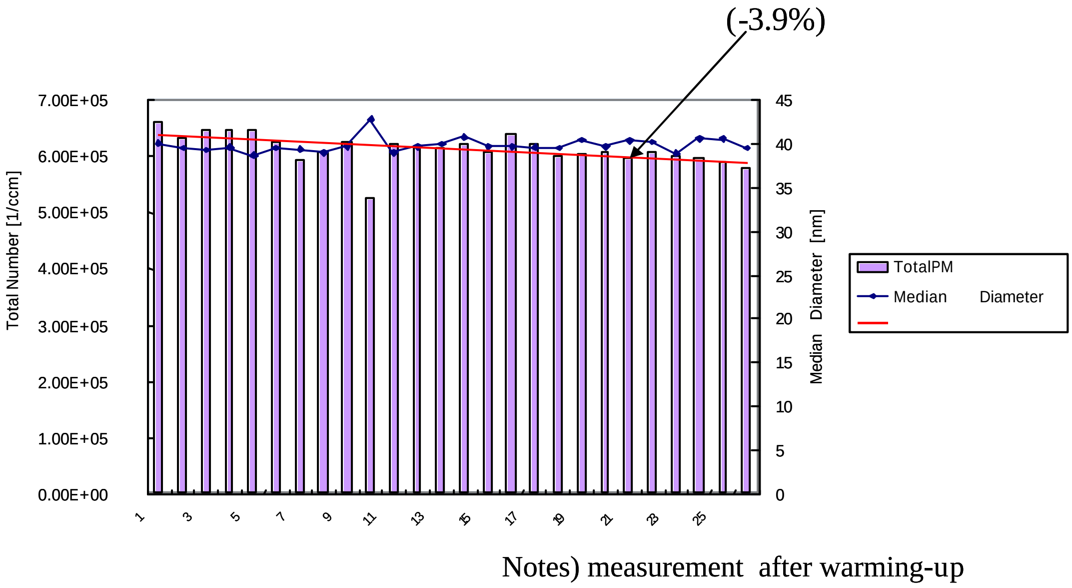
2.2.1 Combustion aerosol standard (CAST)

Currently, the CAST is the only instrument offered as a calibration aid. Particles of a polydisperse distribution are formed in a quenched propane flame. The maximum of the



distribution can vary between 30 and 180 nm (count median diameter CMD) as stated by the manufacturer.

Figure 17a: Repeatability of continuous SMPS-scans (1 to 3 minutes each) with a CAST and a particle distribution with a maximum at 100nm (Source: see annex 5.8, JAMA)



Notes) measurement after warming-up

Figure 17b: Repeatability of continuous SMPS-scans (1 to 3 minutes each) with a CAST and a particle distribution with a maximum at 30nm (Source: see annex 5.8, JAMA)

The particles generated by CAST show a stable average diameter (CMD), but a trend towards decreasing number concentration with time. It appears to be unsuitable as a calibration aid.

2.2.2 Particle surface metric

?? **Diffusion charging sensor (LQ1-DC, Matter Engineering)**

Metric: Active surface [µm^2/cm^3]

The LQ1-DC measurement method uses the probability of deposition of ions on particles for measuring the active particle surface area. Particles are electrically charged by diffusion (unipolar corona charging). The discharge current of the particles is determined by a downstream measurement filter (electrometer). This is a measure for the deposition coefficients of the ions from which the Fuchs surface area or active surface area can be calculated. The measurement does not take place on a single particle basis, only the total active surface area within a defined aerosol volume is given.

The results of surface measurements with LQ1-DC in the nano particle range, can not be explained and are not plausible (see Figure 18).

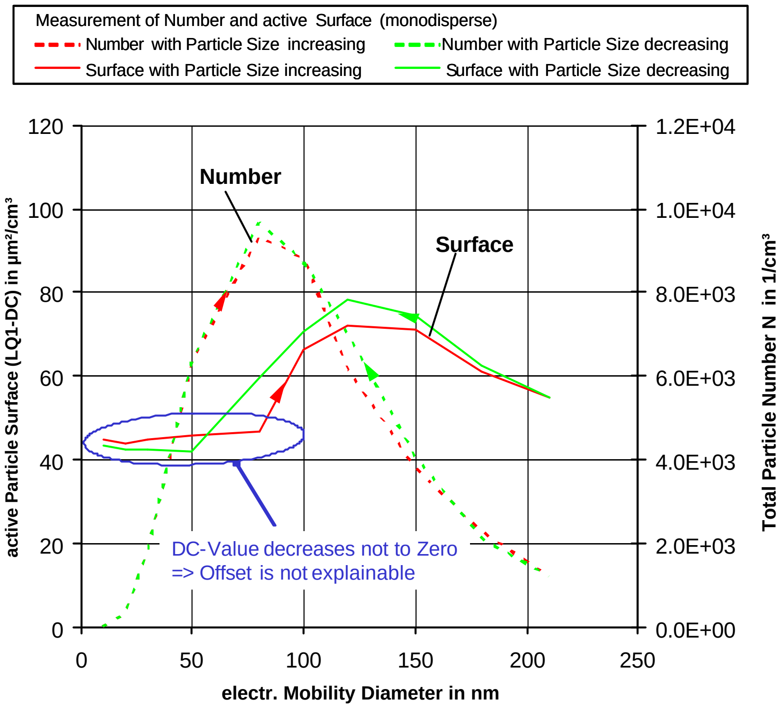


Figure 18: A combination of number (CPC) and surface measurement (LQ1-DC) of monodisperse diesel engine particles, which are sorted by DMA, is shown. The resulting NSD has a maximum at 80 nm as expected. Surface measurement shows a maximum at 120 nm electrical mobility diameter. Below 70 nm there is an offset value, which is not explicable. (Source: See Annex 5.1, ADA).

Measurement of diesel exhaust may lead to contamination of the corona needle (see Figure 19). The principle of charging particles by corona is also used on electrostatic air cleaners. It is known that particle deposits at the corona needle can disturb the discharge. There are hints that this ageing effect can occur also at the LQ1-DC when measuring diesel engine exhaust, although the cause of this has not been established. Changes in particle charging influence the surface measurement directly.

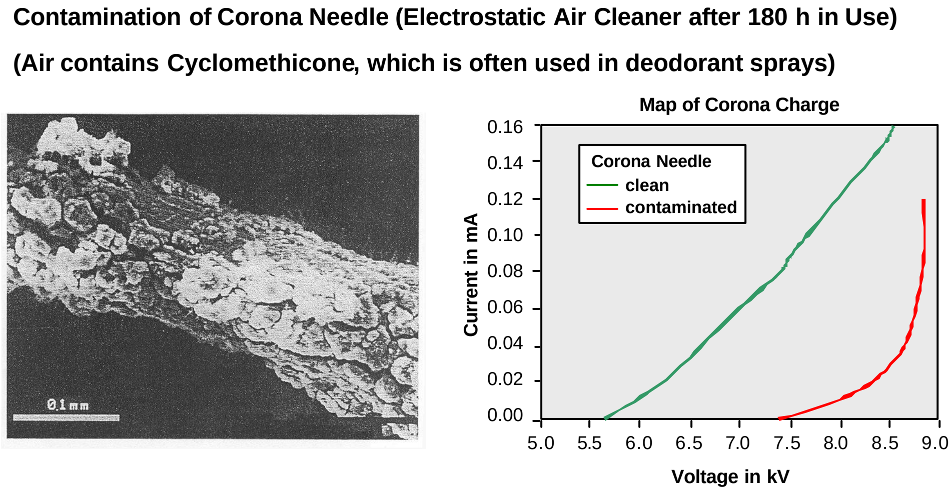


Figure 19: Contamination of Corona Needle (Electrostatic Air Cleaner). (Source: See Annex 5.1, ADA).

Measurements with the LQ1-DC in the particle range less than 70 nm are not plausible. The observed ageing effects make the general suitability of a corona charger questionable. Humidity and variation of the inner electrical insulation caused by deposited particles give a poor stability of the measurement signal. Additionally, no absolute particle surface calibration standard is available for system verification.

?? Photoelectric Aerosol Sensor (PAS 2000; EcoChem)

Metric: Photo electric charging [fA]

The PAS 2000 measurement method makes use of the sensitivity of particles to photoelectric charging. The wavelength of the UV radiator (222 nm) used for the charging is adjusted to suit the ionization of polycyclic aromatic hydrocarbons (PAH) deposited on the particles.

The discharge current of the particles is determined by a downstream measurement filter (electrometer). This can be correlated with the PAH deposited on the particles. With an appropriate calibration, predictions of mass concentration of PAH in µg/m^3 can be made. In some cases this instrument is used for the measurement of Elemental Carbon (EC)[14]. For diesel exhaust measurements, the measured photoelectric signal can not be correlated to the gravimetric mass (see Figure 20).

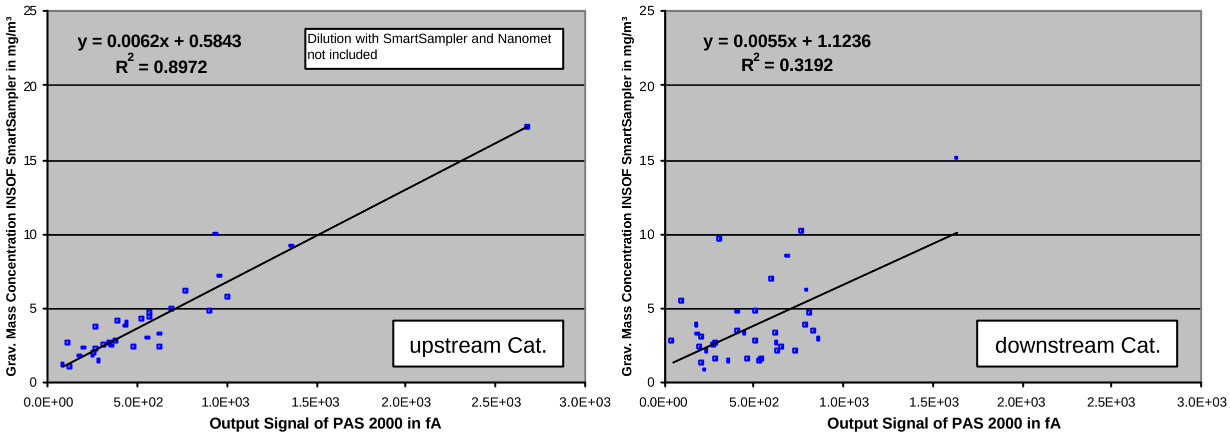


Figure 20: Correlation between gravimetric particulate mass (INSOF) and Output Signals of PAS 2000, separated upstream and downstream of oxidation catalyst. (Source: See Annex 5.1, ADA).

It is observed that the PAS 2000, in principal, is not capable of performing particle surface or mass measurements in exhaust gas from internal combustion engines with reliable and quantitative results. Additionally, no absolute calibration standard is available for system verification.

2.2.3 Particle number metric

?? Electrical Low Pressure Impactor ELPI

Metric: Aerodynamic size distribution of diffusion charged particles [1/cm^3, 12 size bins]

The ELPI method is based on the impactor principle in which the particles are sorted, according to their aerodynamic diameter, into 12 size-classes and are separated on the impactor plates. The counting of the previously electrically charged particles takes place at each stage by means of an electrometer. From the electric charge, assuming an idealized multi-charging based on spherical particles, a complex calculation is

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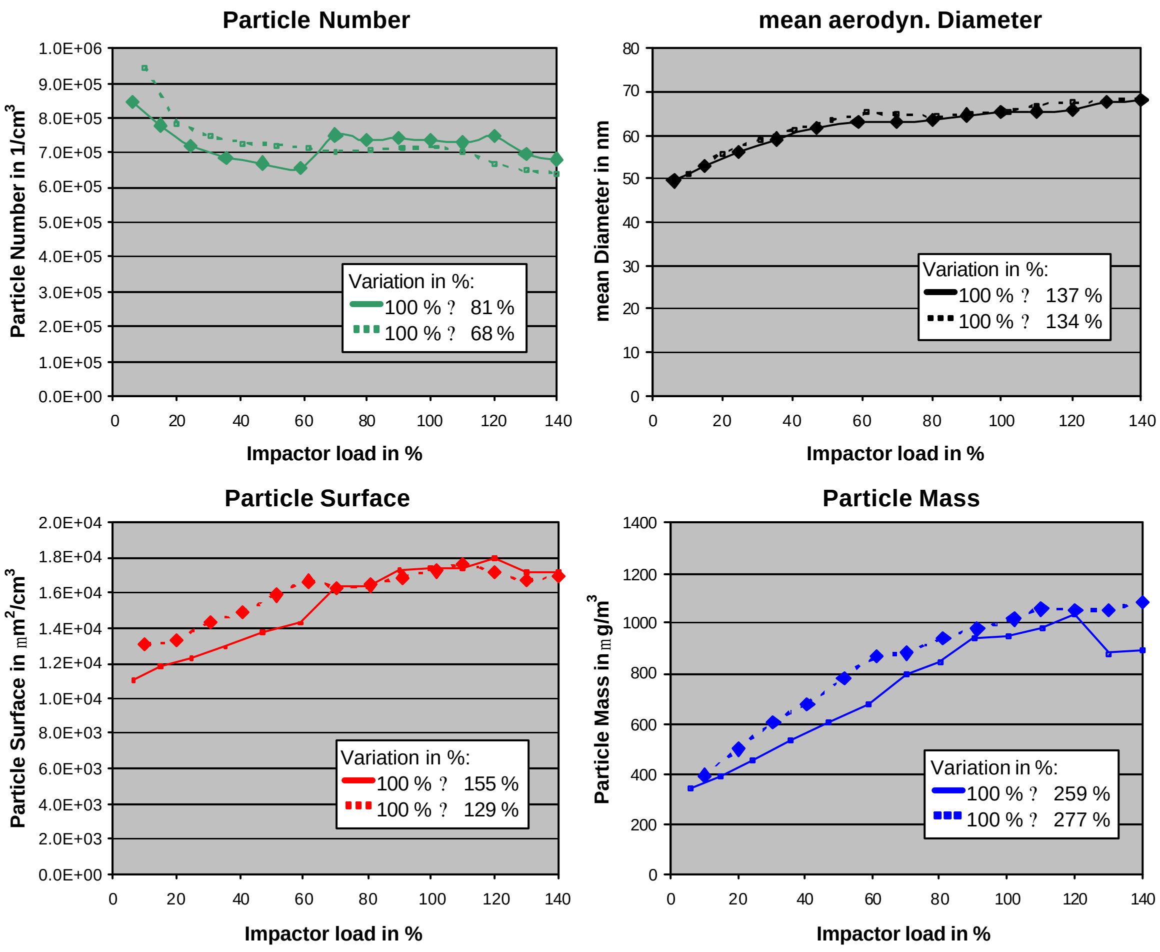
[14] Przybilla; Berkhahn; Burtscher; Dahmann; Matter; Rietschel
"Monitoring diesel particulates in working areas with the photoelectric aerosol sensor"
Gefahrstoffe - Reinhaltung der Luft Nr. 6, June 2002

necessary to convert to particle number. The particles remain on the impactor plates after their detection.

The total number of collected particles is displayed by the device as "Total estimated particle mass load in %" as the sum of all discharge currents. Depending on the impactor load value, the device must be cleaned frequently, i.e. be dismantled to ensure proper functioning.

A long-term test is carried out at a constant engine operating point with continuous measurement of the NSD over a period of 2 - 3 test days. The impactor load value rises from 0 % (cleaned device) to 140 %. The measured NSDs are a function of the value of impactor load and change in the course of the test. The values derived from NSD (number, aerodynamic diameter, surface, mass) depend on the impactor load of the instrument (see Figure 21). In addition the measurements reveal that the changes in the NSD are not reproducible. A correction of ELPI measurements, dependent on the current impactor load, to a defined (e.g. cleaned) reference state of the device is therefore not possible. The steepest gradients occur in the first phase of each set of measurements at around 0 ... 30 % impactor load. This means that cleaning of the device before each individual measurement does not provide a valid solution.



Impactor load value as displayed by ELPI

Figure 21: Particle number, aerodynamic diameter, surface and mass calculated from ELPI measurement with rising impactor load (1st (continuous line) and 2nd measurement (dotted line)). Some of the curves show steps in the course due to breaks (measurement stopped over night) during the measurement period of 3 days. The value of particle concentration at the next morning changed considerably compared

to the value of the day before. The total number of particles measured tend to drop with increasing impactor load whilst the mean measured particle diameter increases. At 140 % impactor load, the total number amounts to 81 % and the mean diameter 137 % of the value at the initial, cleaned state. For the values calculated from these two measured variables for total surface area and total mass (spherical form with uniform density assumed) result in even greater changes: the surface area increases to 155 % of the initial value for 140 % impactor load, the mass increases even more to 259 % of the initial value. The primary cause of this is the change in NSD in the upper ELPI stages where the larger particles are separated off, since these particles make an above-average contribution to the total surface area and total mass. After the device was cleaned, the test was repeated to check the reproducibility of the change in impactor load as a function of time. The changes are reproducible. (Source: See Annex 5.1, ADA)

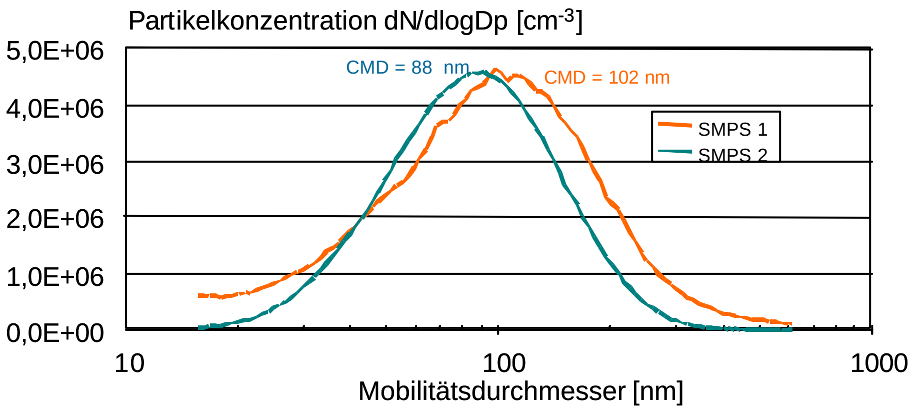
It has not yet been possible to assess the long-term behavior of the corona principle used by ELPI for particle charging. With a Matter-Engineering LQ1-DC instrument, which uses a similar measurement method based on this charging principle, quartz deposits on the corona needle can substantially alter the charging characteristic (see Section 2.2.2).

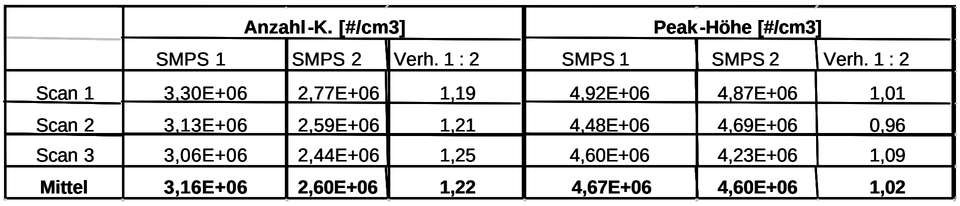
The reproducibility and comparability of ELPI measurements is limited unless a demanding maintenance/operation protocol is strictly observed. The ELPI is not suitable for performing regulatory measurements. Additionally, no absolute particle number calibration standard is available for system verification.

?? Scanning Mobility Particle Sizer SMPS

Metric: Mobility size distribution of particles in the load equilibrium classified by their mobility behaviour in a electric field. Particles counted by an optical single particle counter. [1/cm^3, 64 size bins].

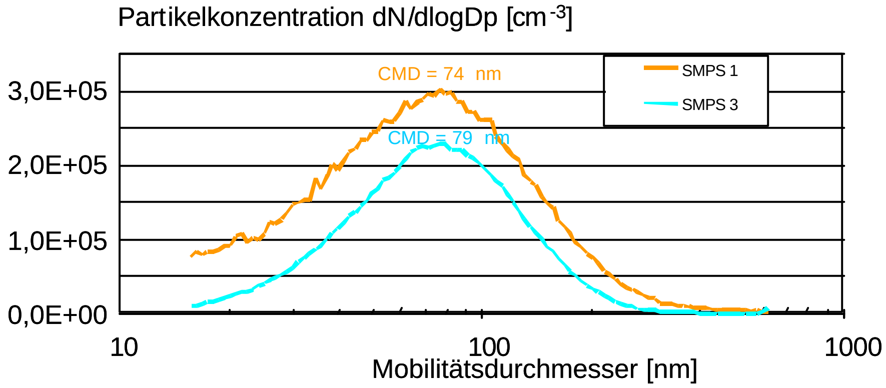
A comparison of three SMPS instruments shows significant differences in the results even though two were used just after calibration by the manufacturer (see Figure 22a/22b). The results show that the basic requirements for the calibration ability and reproducibility are not fulfilled for this type of instrument. Another drawback is that the instrument is not capable of dynamic measurements.





	Anzahl-K. [#/cm3]			**Peak-Höhe [#/cm3]**		
	SMPS 1	SMPS 2	Verh. 1 : 2	SMPS 1	SMPS 2	Verh. 1 : 2
Scan 1	3,30E+06	2,77E+06	1,19	4,92E+06	4,87E+06	1,01
Scan 2	3,13E+06	2,59E+06	1,21	4,48E+06	4,69E+06	0,96
Scan 3	3,06E+06	2,44E+06	1,25	4,60E+06	4,23E+06	1,09
Mittel	**3,16E+06**	**2,60E+06**	**1,22**	**4,67E+06**	**4,60E+06**	**1,02**

Figure 22a: Comparison of SMPS measurement instruments 1 and 2, SMPS1 (Model 3071/3025) and an SMPS2 (Model 3080/3010). SMPS2 was used just after calibration by the manufacturer. SMPS1 showed a median number diameter that was 14 nm larger (15% difference) and a higher total number of approx. 22%. (Source: See Annex 5.6, VW).



	Anzahl-K. [#/cm3]			Peak-Höhe [#/cm3]		
	SMPS 1	SMPS 3	Verh. 1 : 3	SMPS 1	SMPS 3	Verh. 1 : 3
Scan 1	2,099E+05	1,394E+05	1,51	3,108E+05	2,405E+05	1,29
Scan 2	2,166E+05	1,326E+05	1,63	3,083E+05	2,317E+05	1,33
Scan 3	2,181E+05	1,293E+05	1,69	3,157E+05	2,236E+05	1,41
Scan 4	2,165E+05	1,288E+05	1,68	3,060E+05	2,239E+05	1,37
Mittel	**2,153E+05**	**1,325E+05**	**1,62**	**3,102E+05**	**2,299E+05**	**1,35**

Figure 22b: Comparison of SMPS measurement instruments 1 and 3. , SMPS1 (Model 3071/3025) and SMPS3 (Model 3080/3010) showed that the total particulate number was considerably higher as compared to SMPS1 (62 %) and that the median number diameter even deviated in the opposing direction (-5 nm) compared to SMPS3. (Source: See Annex 5.6, VW):

The SMPS does not fulfil basic requirements for quantitative or reproducible measurements. It is not capable of dynamic measurements. Additionally, no absolute particle number calibration standard is available for system verification.

?? Condensation Particle Counter CPC

Metric: Particles enlarged by alcohol condensation counted by an optical single particle counter. [1 to 10^4 particles/cm^3].

Referring to section 2.1 subsection `Nucleation and artefact particle effects`; the CPC is potentially susceptible to large measurement artefacts. However, parallel PM and particle number measurements show (see Figure 23) that, in principle, particle number correlates with mass measurements when the above mentioned effects are avoided (nucleation, artificial particles). However, the gradient of the relationship can not be reproduced because of the dependency of the particle number and morphology on the measurement conditions.

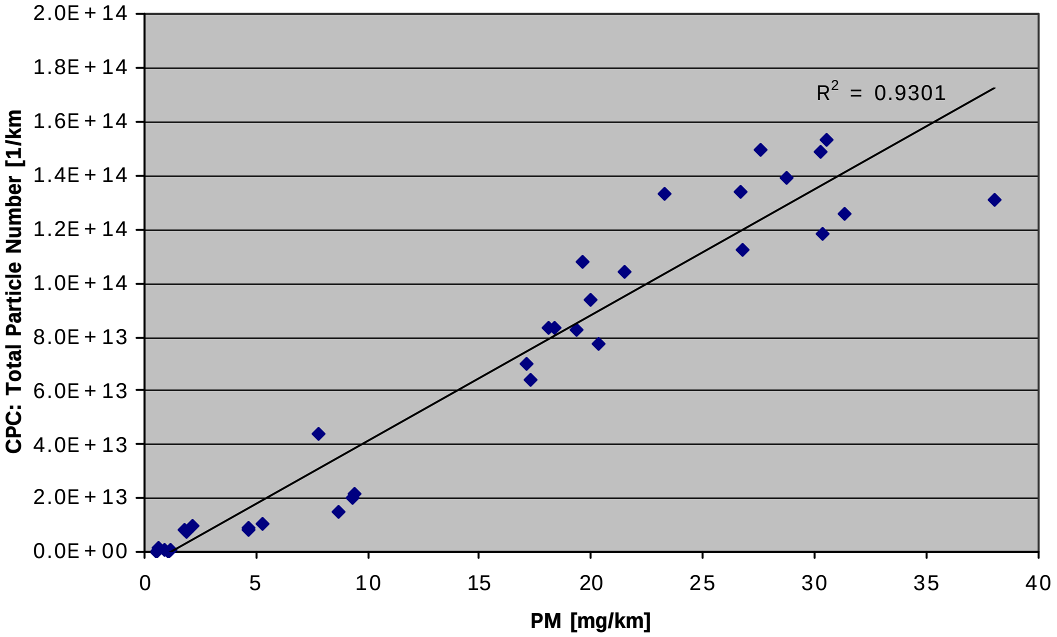


Figure 23: Correlation between CPC and PM measurements for a gasoline direct injection vehicle and a diesel vehicle in the NEDC transient cycle and at constant speeds of 50 km/h, 100 km/h and 120 km/h. The tests were conducted at a roller dyno test stand. For the CPC measurements, the sample was treated by using secondary dilution of 100 to 800 times with a rotating disc diluter after the CVS dilution tunnel. (Source: VW)

Compared with parallel PM measurements the CPC shows higher relative standard deviations at the same emission level as can be seen in Figure 24.

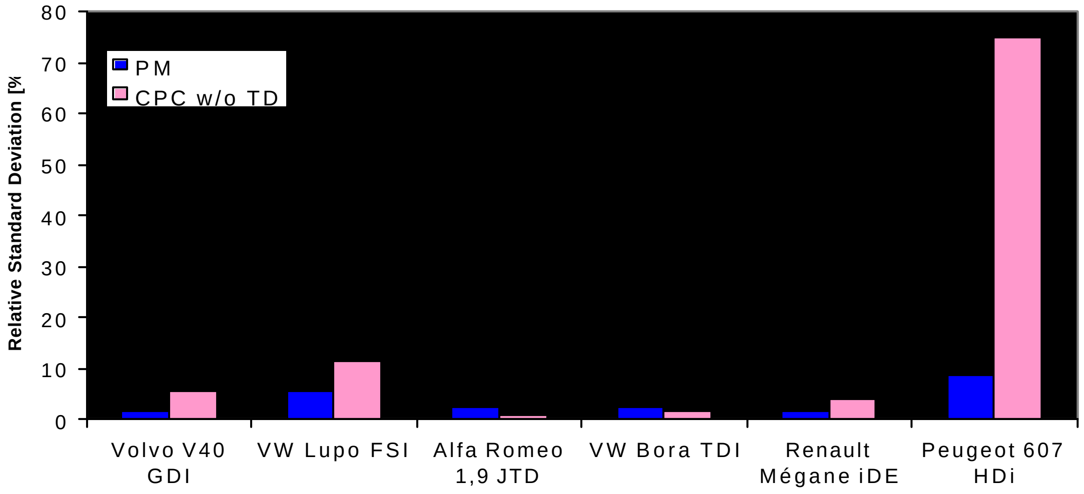


Figure 24: Comparison of relative standard deviation of parallel PM and CPC measurements in the NEDC driving cycle on a roller dyno. The CPC measurements were carried out by sampling out of the CVS tunnel and additional dilution by a factor of 10 with a ejector diluter.
At low particulate emission conditions, the CPC measurements are less reproducible than the PM measurements. (Source: ACEA Programme on the Emissions of Fine Particles from Passenger Cars [2], www.acea.be)

The CPC is very sensitive to nucleation effects. Due to the lack of size resolved information, these effects are not identifiable. The operation range is limited (variable high dilution needed) and the repeatability is low compared to state of the art PM mass methodology. Additionally, no absolute particle number calibration standard is available for system verification.

2.2.4 Particle mass metric

?? Photo acoustic soot sensor – PASS

?? Laser induced Incandescence – LI^{2}SA

Metric of PASS: Photo acoustic response of soot particles stimulated by light absorption.

Metric of LI^{2}SA: Incandescence of laser pulse-heated soot.

Time-resolved measurements of elemental carbon are feasible with the photo acoustic sensor as well as with the laser induced incandescence system. Neither technique is susceptible to nucleation particles. No cross interference of other exhaust gas components is exhibited.

Both systems reveal a very good correlation with PM mass. Figures 25, 26 and 27 show the correlation characteristics of the PASS and LI^{2}SA measurements with the standard gravimetric method or thermogravimetric results (non volatile particulate mass on filter) respectively. As LI^{2}SA, PASS and Coulometry are soot-specific measurement tools, they account for typically about 80 % of total gravimetric mass. Results from opacimetry and thermogravimetry are slightly higher and reach values around 90 %.

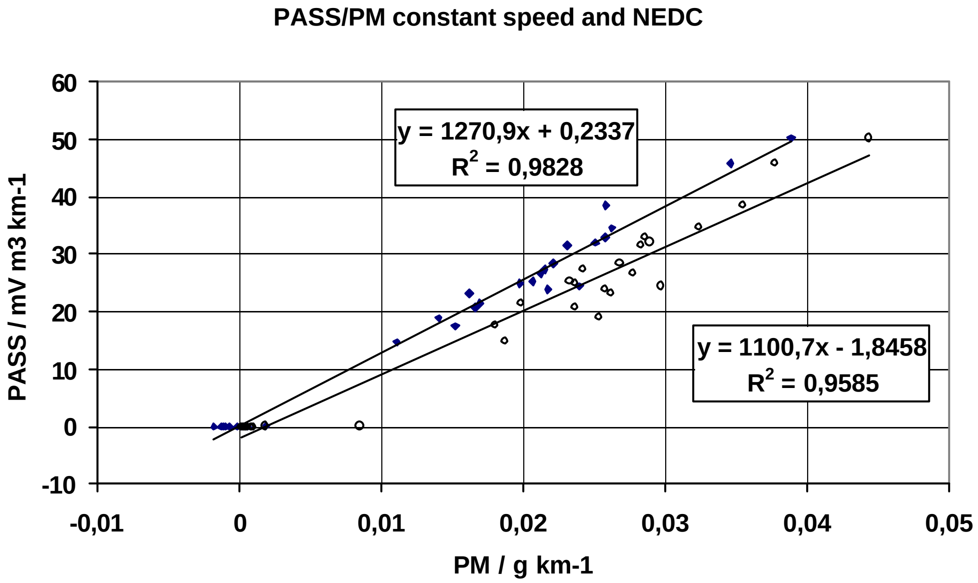


Figure 25: Correlation of integrated PASS signal and particulate mass (open circles) measured during constant speed tests and NEDC2000 for 1 diesel passenger car and 1 diesel passenger car equipped with a particle trap. The correlation is further improved (filled rhombus), if volatile organics and soluble inorganic material are subtracted from the measured PM mass. (Source See Annex 5.5, Ford)

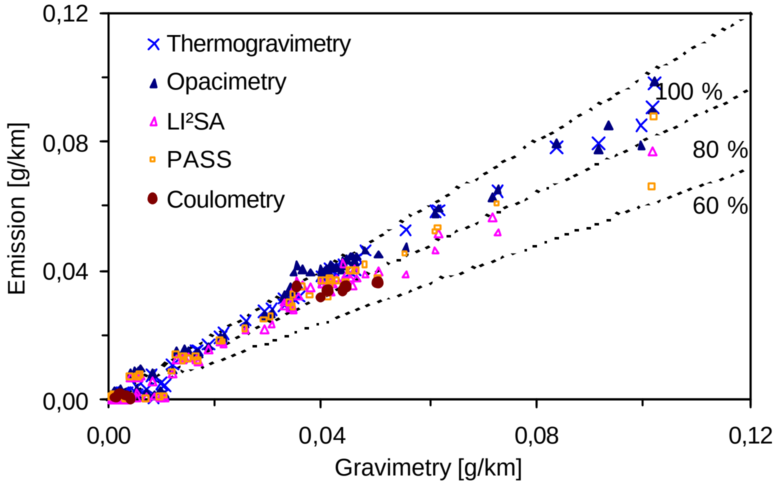


Figure 26: NEDC-Phase average concentrations of different mass-based measurement methods of different diesel and gasoline vehicles plotted over results from gravimetry. Opacimetry: Opacity across the CVS tunnel. Coulometry: Particle load of filters analysed by coulometry for total carbon mass. (Source: See Annex 5.4, DaimlerChrysler)

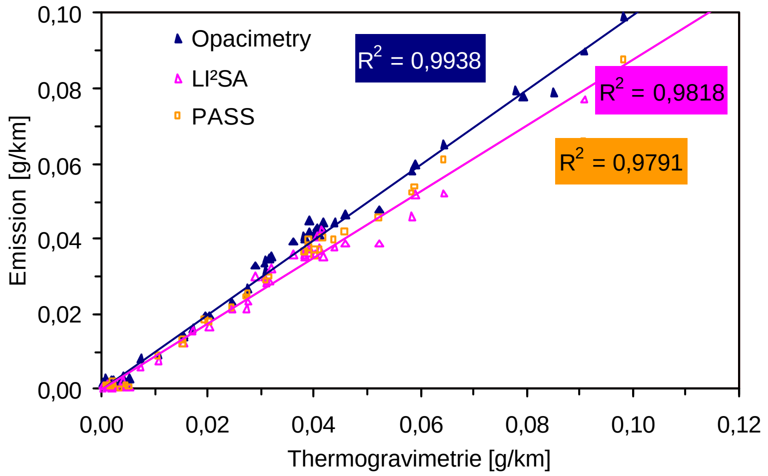


Figure 27: Correlation of mass calculated from opacity, LI²SA and PASS with thermogravimetric data of different diesel and gasoline vehicles in the NEDC (Source: See Annex 5.4, DaimlerChrysler)

At very low emission levels PASS and LI²SA show a reasonable correlation (see. Figure 28). These results confirm that PASS and LI²SA are sensitive enough also for the measurement of future emission levels.

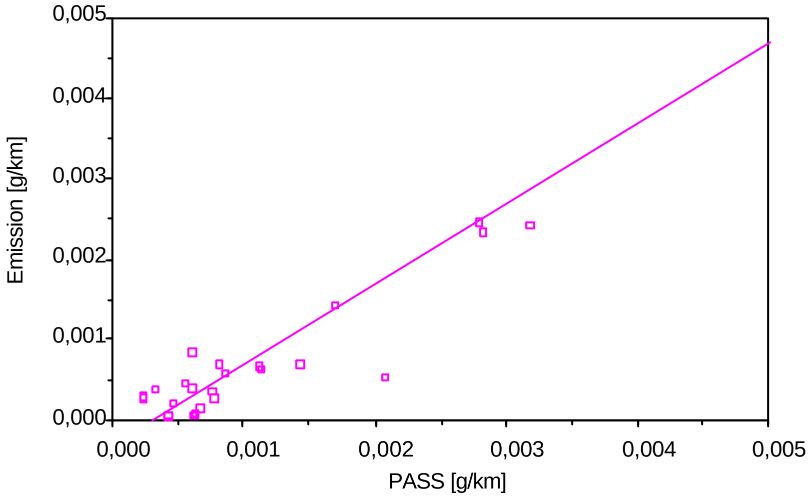


Figure 28: Correlation of LI^{2}SA with PASS at low emission levels (Source: See Annex 5.4, DaimlerChrysler)

The time resolution of PASS and LI^{2}SA (see Figure 29) is satisfactory – which makes these systems suitable for real time measurement purposes. The PASS offers a sampling rate in the order of 3 Hz, the LI^{2}SA system featured a sampling rate of 0.5 Hz, (systems with a sampling rate in the order of 20 Hz are available), for the examined measurement set up used (sample extraction from CVS tunnel, with a delay time depending on length of sample line).

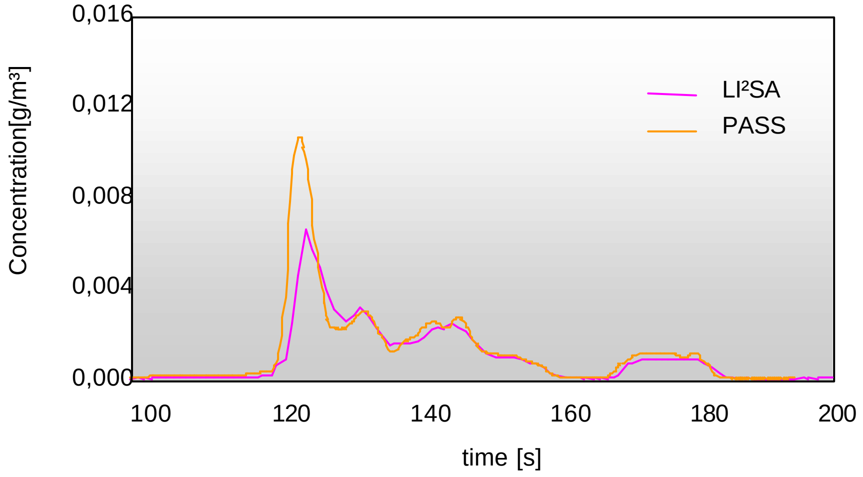


Figure 29: Comparison of time resolution (100 s section of an NEDC). (Source: See Annex 5.4, DaimlerChrysler)

The measurement techniques, PASS and LI^{2}SA, for the determination of the mass emission of elemental carbon are characterised by the following:

- Significant correlation to gravimetrically determined solid particle emissions (regression coefficient = 0,98).
- Cross interference to other exhaust components (e.g. volatile nanoparticles) does not occur.
- With a time resolution of ? 5 Hz it is suitable for real time measurements.

Measurement of elemental carbon by PASS or LI^{2}SA provides a sensitive and time-resolved measurement technique, which could in principle be absolutely calibrated by coulometry.

Additionally the LI^{2}SA measurement technique offers the possibility to determine the primary particle diameter of the soot particles. As Figure 30 shows, this diameter is independent of particle concentration (and thus engine load) and combustion principle. For all vehicles investigated, the primary particle size was in the range of 25 – 35 nm.

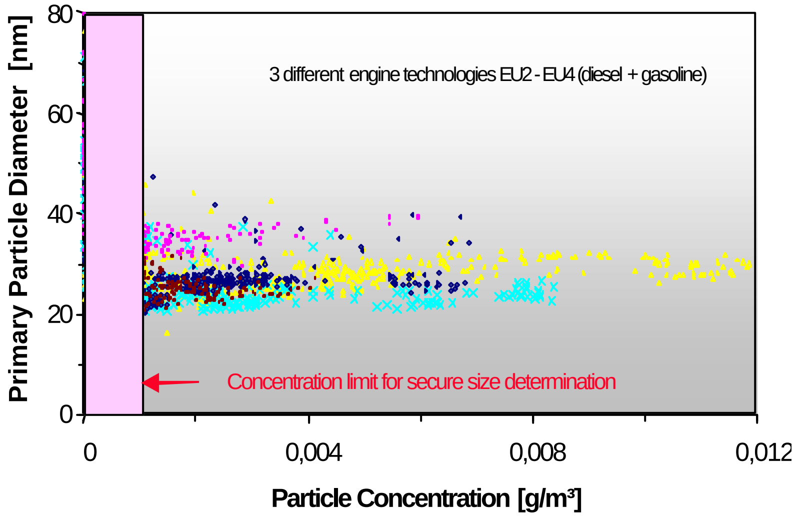


Figure 30: Primary particle size plotted over particle concentration for four different vehicles. The primary particle size is independent of particle concentration and combustion principle. (Source: See Annex 5.4, DC)

The measurement of primary particle diameter by LI^{2}SA does not offer notable size differences, therefore the usefulness of this parameter is questionable.

?? Dekati Mass Monitor MasMo

The operation principle of the newly developed MasMo is based on particle charging, particle mobility measurement, the assumption of a unimodal symmetrical size distribution, particle size classification with inertial impaction (6 stages: 0-1.2 µm), and electrical detection of charged particles for real time measurement of particle mass.

The missing link to calculate the particle mass from size resolved aerosol measurements is the particle density. Dekati solved the problem by a combination of a ELPI and a simple SMPS. Comparing the aerodynamic diameter with particle mobility information should make it possible to calculate an average particle density.

VW tested a prototype version under different conditions. The data analysis reveals that for Diesel vehicle measurements the MasMo results correlate well with the gravimetric results. Gasoline and gasoline direct injection prototype vehicles results correlate well but show a significant discrepancy in the slope (see Figure 31). The reasons for this discrepancy is probably due to the limited ability to measure the correct particle density.

In addition a significant scattering in the MasMo result was observed which reveals that the MasMo does not achieve the reproducibility of the PM mass measurements.

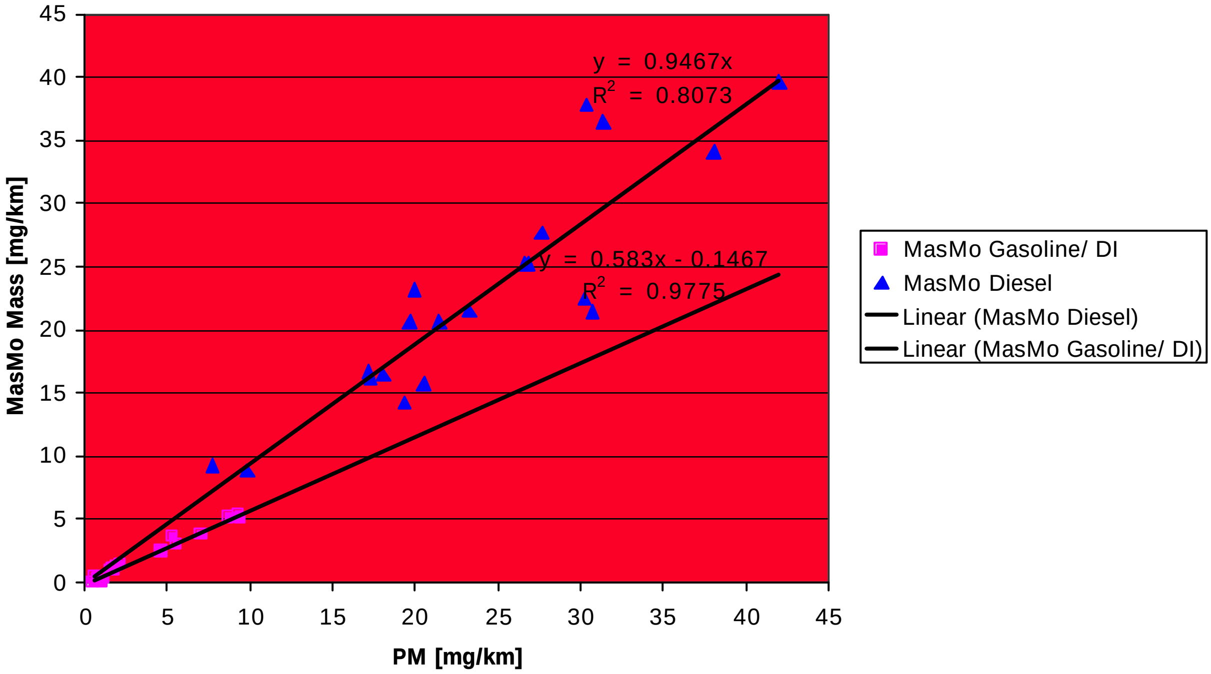


Figure 31: Correlation between MasMo and PM for Diesel, gasoline and gasoline direct injection vehicles in the NEDC transient cycle and at constant speeds of 50 km/h, 100 km/h and 120 km/h. The tests were conducted on a roller dyno test stand. For the MasMo measurements the sample was treated by using secondary dilution of 10 with a Dekati ejector diluter after the CVS dilution tunnel. (Source: VW)

The general applicability of the MasMo for regulatory engine exhaust measurements is questionable. In its current development status, it has low reproducibility compared to state of the art PM methodology. In addition, no absolute particle density calibration standard is available for system verification.

2.2.5 Instrumentation correlation assessment

A study was carried out to assess the correlation between several instruments of the same type simultaneously, using CAST and diesel exhaust as the particle source. This study included testing different types of instruments (DMA+CPC, ELPI, DC).

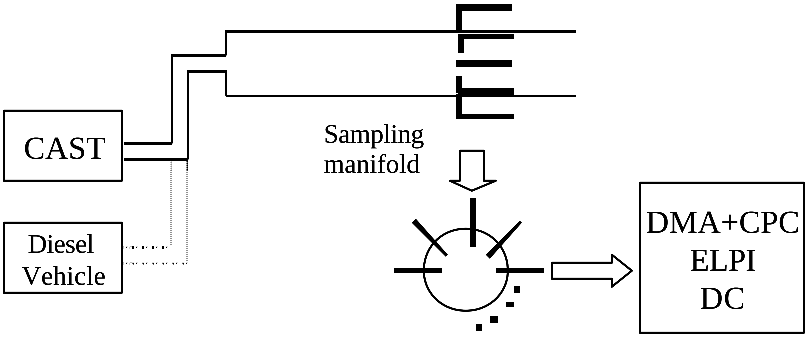


Figure 32: Correlation study test set-up (source: JAMA, annex 5.8)

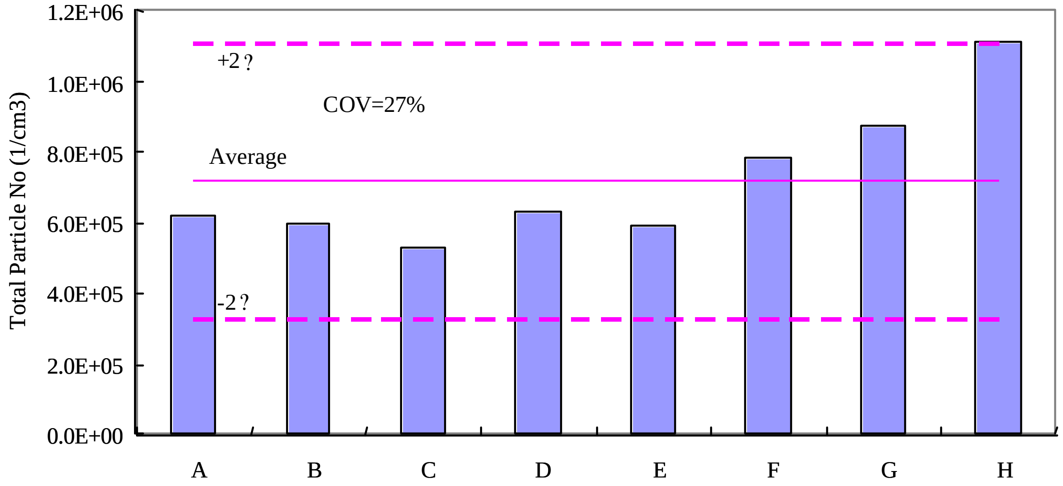


Figure 33a: Simultaneous measurements of total number of particles from CAST (CMD = 100 nm) using eight ELPIs. COV = Coefficient of variation (source: JAMA, annex 5.8)

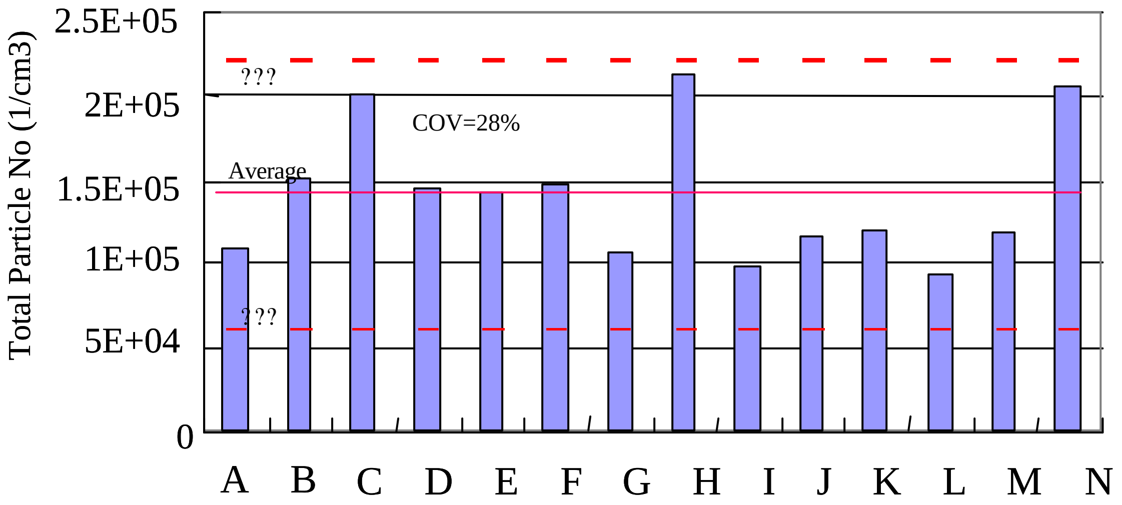


Figure 33b: Simultaneous measurements of total number of particles from CAST (CMD = 100 nm) using 14 DMA+CPCs. COV = Coefficient of variation (source: JAMA, annex 5.8)

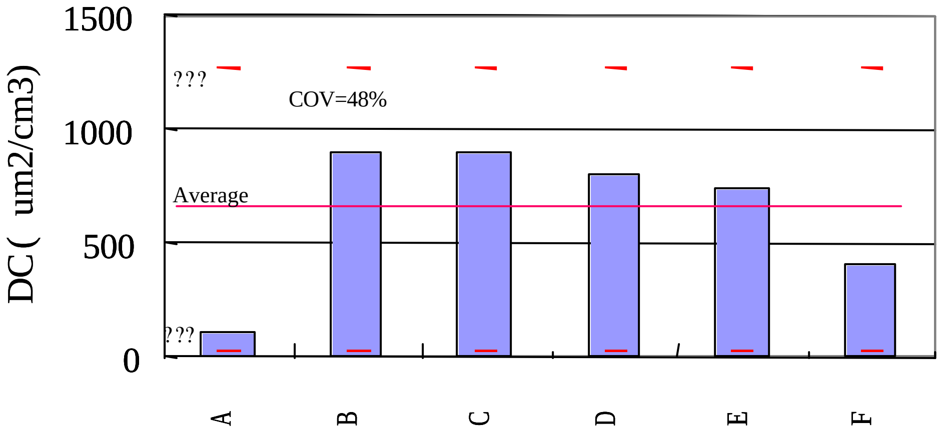


Figure 33c: Simultaneous measurements of total number of particles from CAST (CMD = 100 nm) using 6 DCs. COV = Coefficient of variation (source: JAMA, annex 5.8)

	CAST(1) MP3(100nm)	CAST(2) MP6(30nm)	Diesel Exhaust
ELPI Total Particle Number	27%	40%	23%
DMA+CPC Total Particle Number	28%	19%	27%
DC Total Surface Area	48%	48%	54%

Figure 34: Summary of correlation tests (COV) with different kinds of equipment for particle measurements (DMA+CPC, ELPI, DC) using CAST and diesel exhaust (source: JAMA, annex 5.8).

The variation between several instruments of the same type is very high even when they are simultaneously measuring the same particle source. Since a robust calibration method does not exist, these instruments are clearly not suitable for certification.

2.3 Evaluation of the gravimetric method

2.3.1 Light duty

The particulate gravimetric measurement method is described in EU regulation 70/220/EC. For the measurement of the particulate mass, a sample of the diluted exhaust gas (partial flow) is extracted from the dilution tunnel and drawn through a filter system.

The particulate mass is calculated knowing the mass of particulate on the filter, the partial flow volume and the total flow volume. The higher boiling point hydrocarbons and sulphur compounds are also deposited on the filter. As a result of the reduction in hydrocarbon emissions and the use of fuels with a low sulphur content, the proportion of those components which can influence the particulate measurement is reduced.

The detection limit of the current standard gravimetric procedure for light duty vehicles was examined by DaimlerChrysler, BMW and Volkswagen.

In order to determine the limit of detection of the entire process (weighing, filter handling, loading, weighing) NEDC-blank tests (tests without vehicle) were performed by DaimlerChrysler (see Figure 35). As a result, the LOD (3 * ?) is estimated to be 0.025 mg. This is equivalent to approximately 1 mg/km in an NEDC which is 4 % of the Euro-IV emission limit. By further optimization steps of the gravimetric method (e.g. optimized flow, micro balance with increased accuracy) it will be possible to decrease the LOD to approximately 0.01 mg filter loading.

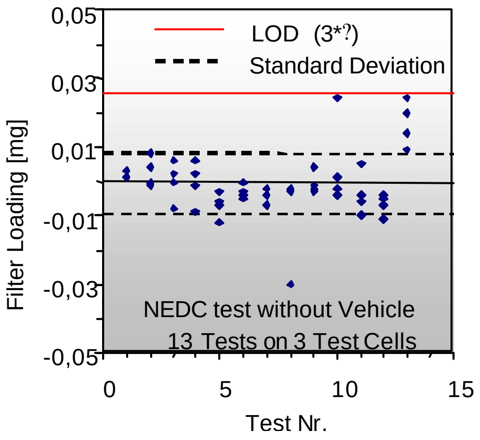


Figure 35: Determination of LOD of gravimetric method by blank tests. The difference in weighing before and after test is shown for NEDC tests without a vehicle. The zero scatter (standard deviation ?) is +/- 0,008 mg. Therefore, the LOD (3 * ?) is estimated to be 0,025 mg filter loading (source: see annex 5.4, DaimlerChrysler).

To estimate the lower limit of the gravimetric mass measurement, BMW also carried out NEDC blank-tests. The results are shown in Figure 36.

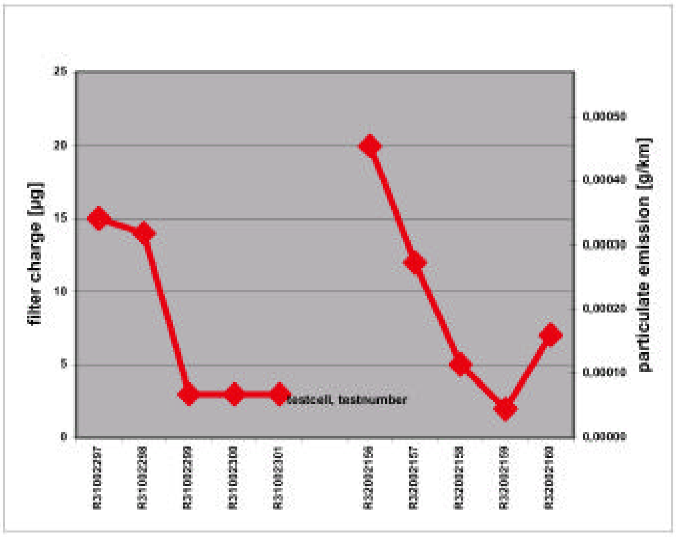


Figure 36: Results of blank-tests on two chassis dynos. Instead of sampling the vehicle exhaust, the air of the test-cell was sampled during a NEDC test. (Source: See Annex 5.2, BMW)

Five NEDC blank tests were made in succession on each of two chassis dynos. The results show that the mass emission appears to decrease with increasing test number. It was assumed that particles are removed from the surface of the sampling system and in time this “particle-source” decreases.

The mean value of all 10 tests is 0.0002 g/km with a standard deviation of 0.00015 g/km. One contribution to this relatively high standard deviation is from the decreasing “particle-source”. Compared with the Euro-IV limit of 0.025 g/km this standard deviation of 0.00015 g/km is less than 1% of that limit value. Tests with very low emission vehicles had smaller standard deviations of about 0.0001 g/km.

Volkswagen investigated the detection limit of the particle measurement procedure by conducting NEDC blank tests and additionally NEDC test with vehicles with a particulate emission level less than 0,010 g/km. As DaimlerChrysler and BMW did, the determination of the “detection limit” of the complete measurement sequence was carried out without using a vehicle, but sucking ambient air into the CVS system. As shown in Figure 37, the “background emissions” value was below 0.001 g/km. The corresponding mean filter loading during the determination of the background value was 0,02 mg. This is considerably lower than the filter loading of at least 1 mg recommended in the European regulation.

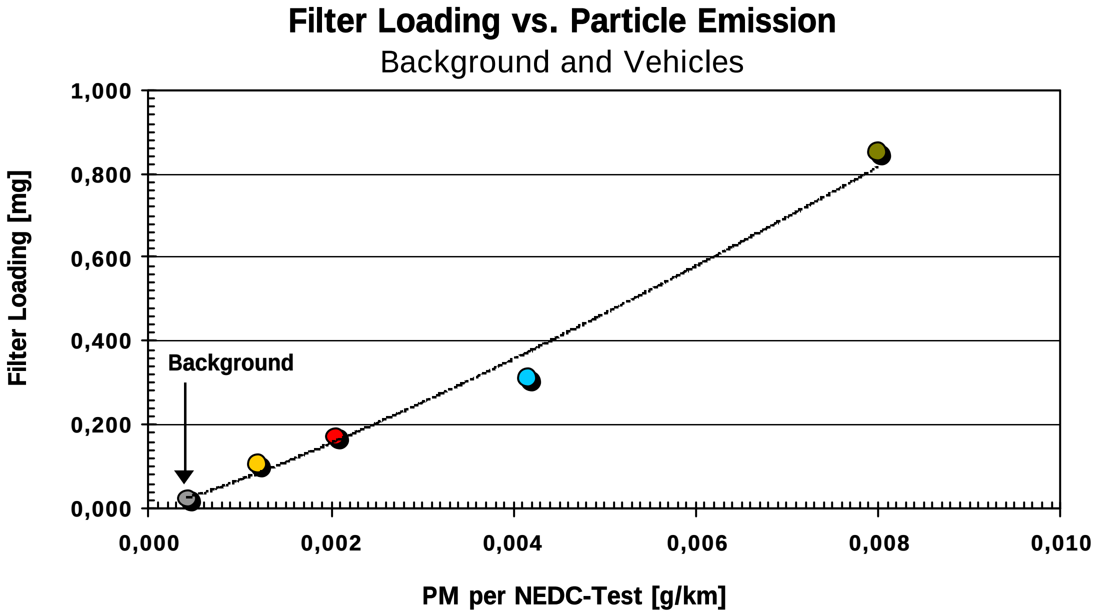


Figure 37: Lower practical limit of the gravimetric measurement procedure. For the determination of the "detection limit" of the complete measurement sequence, NEDC blank tests were performed sucking ambient air into the CVS system. The "tunnel background" value was below 0.001 g/km. The other measurement points show the mean filter loading of the vehicle in the new European Driving Cycle (NEDC). Each measurement point represents a mean value of 3 – 25 measurements. (Source: Annex 5.6, VW)

The measurement sequence with the four vehicles demonstrates a slight scattering of the measurement values (see Figure 38). Their absolute standard deviation can be compared with the standard deviation of the background emissions although the measurement chain has been extended by the parameters chassis dyno, vehicle and driver. The results show clearly that the accuracy of the particulate measurement technique is much better than can be assumed on the basis of the current regulations.

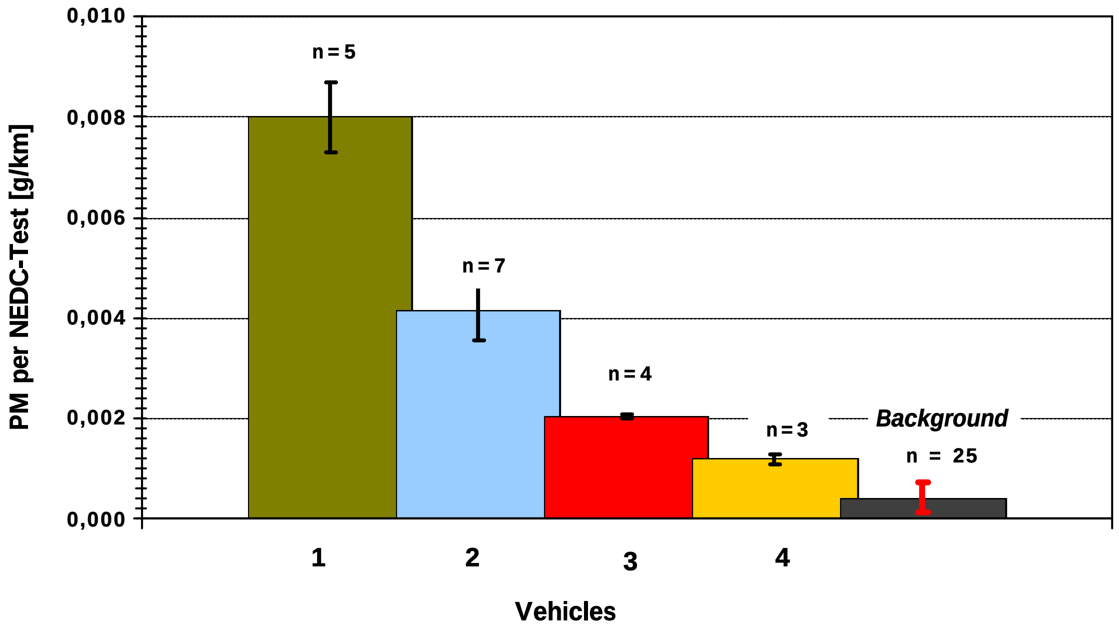


Figure 38: It describes the average particulate emissions in g/km and the standard deviation of four vehicles and the tunnel background measurement. The emissions of 0,0004 g/km (= 0,02 mg filter loading) could be measured with a standard deviation of 0,0003 g/km. This gives a relative standard deviation of approx. 75%. Based on the Euro IV limit of 0,025 g/km, this corresponds with a deviation of the measurement results of less than 2 % for the sampling system including the determination of the volumes and weighting of the filters. The relative standard deviation of the vehicles is between 5 % and 9 %. (Source: See Annex 5.6, VW)

In summary, it can be seen that the gravimetric particulate measurement is a calibratable particulate measurement technique. The boundaries of the particulate measurement process have not yet been reached and further optimisation is possible. Examples include, but are not limited to: determination of stricter tolerances, reduction of the minimum filter loading, sampling only on one filter and conditioning of the dilution air (synthetic…

The standard gravimetric procedure reveals the potential for particulate emission measurements also beyond Euro 4 regarding accuracy and the limit of detection. Optimization possibilities exist for further improvements.

2.3.2 Heavy duty

For heavy-duty engines, the gravimetric method is described in Directive 1999/96/EC and in the US Federal Register Part 86, Subpart N. The measurement principle is identical to the light duty procedure with some minor differences in the details. Those are mainly the widespread use of 70 mm diameter filters and the double dilution system, in which the PM sample from the primary dilution tunnel is diluted once again. The Euro 4 limit values are 0.02 g/kWh and 0.03 g/kWh on the ESC and ETC test cycle, respectively.

PM measurement variability is influenced by two major factors

- PM calculation by the tolerances of the relevant measuring instruments
- PM composition by the sampling conditions and the dilution process

??PM calculation

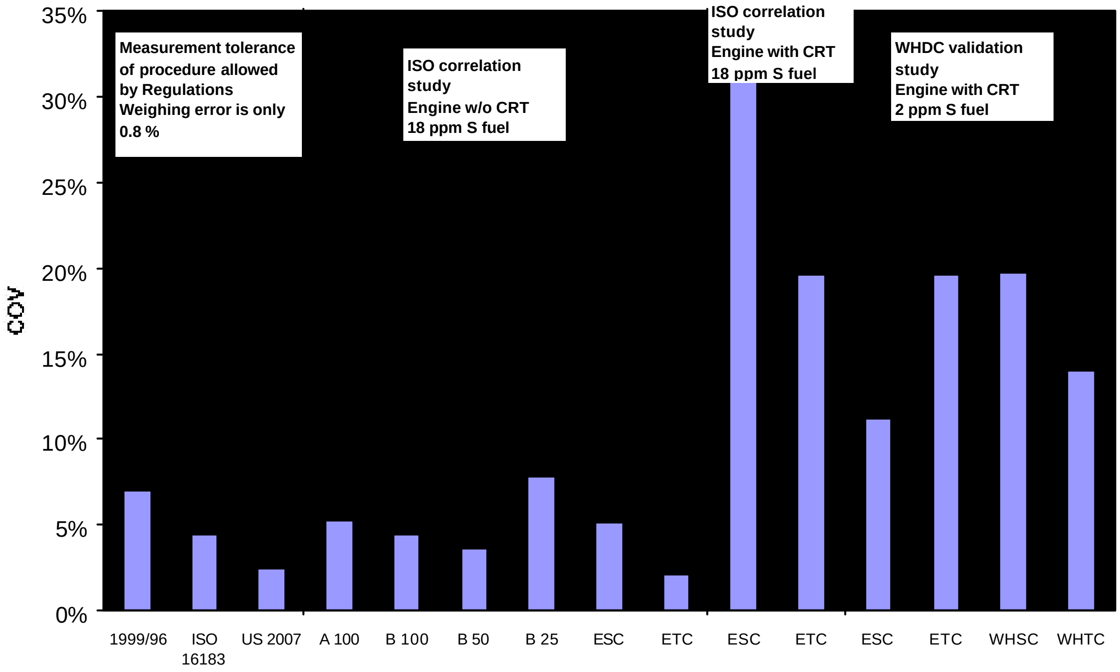
The final PM result of g/kWh is calculated from the differential weighing before and after test, the CVS (or exhaust) flow, the sample flow, and the engine power. The Directive specifies the tolerances of the respective measuring instruments. Figure 39 summarizes the allowable tolerances in comparison to results from round robin testing. It should be noted that the weighing tolerance refers to the tolerance of reference filter weighing rather then to the capability of the balance. The error of a 1 µg balance would be as low as 0.8 % at a 0.25 mg filter loading.

ESC		
Measured Value	Error	Remarks
Particle weight (mg)	? 3.9 %	Based on 2.5 mg particulate weight
Diluted exhaust flow (kg/h)	? 4.0 %	Max. error allowed
Sample flow (kg)	? 2.0 %	
Engine power (kW)	? 2.0 %	Max. allowed error is ? 3.6 %
PM (g/h)	? 5.9 %	average total error (RMS method)
PM (g/kWh)	? 6.3 %	
PM (g/kWh)	4.7 % - 15.8 %	COV range on ACEA round robin
ETC		
Measured Value	Error	Remarks
Particle weight (mg)	? 4.2 %	Based on separate weighing
CVS flow (kg)	? 2.0 %	Max. error allowed
Sample flow (kg)	? 4.9 %	Based on double dilution
Engine cycle work (kWh)	? 2.0 %	
PM (g/Test)	? 6.8 %	average total error (RMS method)
PM (g/kWh)	? 7.0 %	
PM (g/kWh)	3.8 % - 12.0 %	COV range on ACEA round robin

Figure 39: Comparison of theoretical vs. observed measurement variability (Source: ISO TC 22/SC5 engine tests)

The calculated average total error should in principal be representative of the test repeatability of a given lab, expressed as the coefficient of variation (COV = STD/AVE). This has been confirmed by the ACEA round robin, although some labs had significantly worse repeatabilities. Therefore, it can be concluded that test repeatability can be improved if the measurement tolerances of the legislation are tightened.

USEPA in their MY 2007 rules and ISO 16183 haven already taken steps to reduce the tolerances. The potential improvement of measurement variability is shown in Figure 40. Compared to the Directive 1999/96, the EPA procedure is expected to halve the variability. Figure 40 also shows the good agreement between theoretical measurement error and test repeatability (COV) at around 5 %, as found on the ISO correlation study for a typical Euro 3 engine, and thus confirms the results of the ACEA round robin indicated in Figure 39.



<u>Figure 40:</u> Relative PM measurement variability for heavy-duty engines (Source: ISO TC 22/SC5 engine tests)

Test repeatability significantly increases to values between 20% and 30% when the engine is equipped with a particulate trap and operated on a fuel with 18 ppm sulfur content. However, the major reason for this deterioration is not related to the measurement equipment or the low PM values of a trap-equipped engine, but to changes in the PM composition, as outlined in more detail, below. An improvement of the repeatability has been achieved when further reducing the sulfur level to approximately zero.

?? **PM composition**

PM mainly consists of elemental carbon (EC, soot), the volatile organic fraction (VOF) adsorbed onto the soot, and sulfate. Whereas EC is not affected by the dilution process and only slightly affected by the sampling conditions, both VOF and sulfate are very sensitive to dilution and sampling.

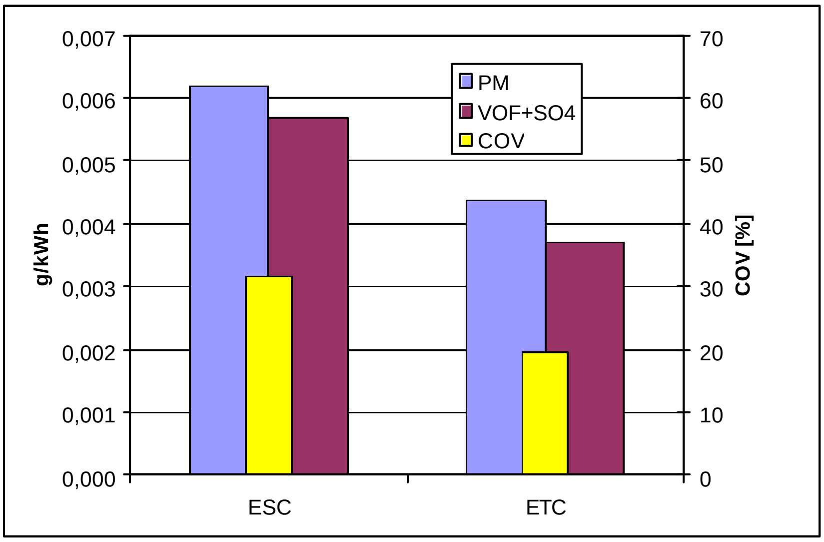


Figure 41: Influence of PM composition on measurement variability for an engine with particulate trap (Source: ISO TC 22/SC5 engine tests).

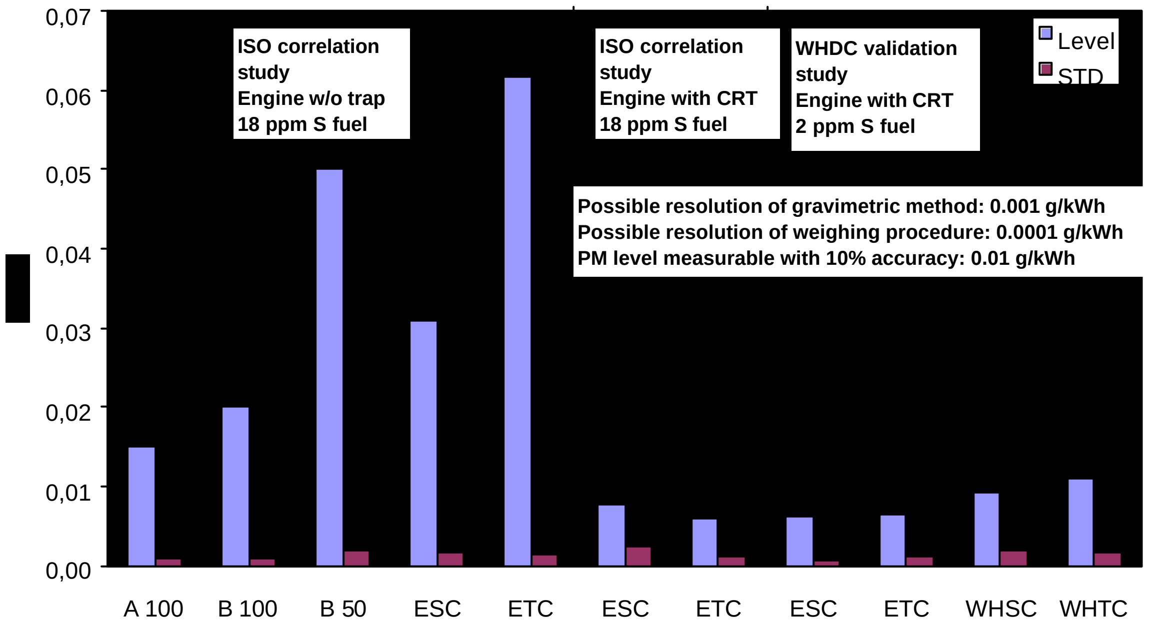
Figure 41 shows the influence of the PM composition on the measurement variability shown in Figure 40 for the trap-equipped engine running with 18 ppm sulfur fuel. It demonstrates that the measurement variability (COV) deteriorates significantly, if sulfate and VOF levels are high and the predominant portion of total PM. Although on both ESC and ETC test cycles the VOF/Sulfate fraction of the total PM is about the same (92% vs. 85%), the COV is more affected by the absolute level, which is 54% higher for the ESC.

As a conclusion, the measurement variability caused by PM composition is related to the PM definition rather than to the gravimetric procedure and will likely occur with any other PM measurement method, as demonstrated in this report. Sulfate formation is a problem specific to exhaust aftertreatment systems with oxidation catalysts. In such a case, high PM variability can only be avoided if sulfur free fuel is used.

??Potential of the gravimetric PM measurement

The potential of the gravimetric method is shown in Figure 42, as derived from the ISO correlation and the WHDC validation studies. Overall, the measured standard deviation (STD) varied between 0.001 and 0.002 g/kWh. With the improvements introduced by US 2007 rules and ISO 16183, the STD can be cut down to 0.001 g/kWh even at very low PM levels, whereby the weighing procedure itself is better by one order of magnitude (0.0001 g/kWh). Such low number is hardly achievable by any other instrument available, today.

For most test laboratories, a STD of 10 % is acceptable at Euro 4 and US 2007 PM levels, although the target value is preferred to be 5 %. Under those assumptions, PM measurement with 10 % accuracy is possible down to a PM level of 0.01 g/kWh. Further improvements towards the 5 % target value are feasible, if the engine is operated with virtually sulfur-free fuel (less than 5 ppm).



Possible resolution of gravimetric method: 0.001 g/kWh
Possible resolution of weighing procedure: 0.0001 g/kWh
PM level measurable with 10% accuracy: 0.01 g/kWh

Fig. 42: Potential of gravimetric PM measurement method for heavy-duty engines (Source: ISO TC 22/SC5 engine tests)

PM measurement is possible down to a level of 0.01 g/kWh with a 10 % measurement error, which is acceptable to most test labs. Further improvements are feasible, if sulfur free fuel can be used. Taking into account this potential of the current type approval method, it should be retained for the future.

3 Discussion

The potential of a measurement instrument for certification testing can be evaluated by comparing it with the following requirements.

TABLE 1
Minimum requirements for the instrument incl. sampling system for measurement of particulate mass, number and surface area

	Mass	**Number**	**Surface**
Measurement Capability	10% of regulatory limit value	10% of CVS dilution air background	Unknown
Calibration checkable on site	yes		
Repeatability	10* % of regulatory limit value		
Reproducibility	20* % of regulatory limit value*		
Time resolution	Integral individual phases of the test cycle		

*Value considers light duty and heavy duty requirements

Mass measurement instrumentation evaluation

nstrument	**Practical**	**Repeatability**	**Reproduci-**	**Time**	**Calibration**

	limit		bility	resolution	checkable on site
ass	Yes, but only EC	Yes	Yes	Yes	Yes, indirectly
i^{2}SA	Yes, but only EC	Yes	Yes	Yes	Yes, indirectly
asMo	Yes	limited	limited	Yes	No
ravimetry	Yes	Yes	Yes	Yes	Yes

Number measurement instrumentation evaluation

nstrument	Practical limit	Repeatability	Reproduci-bility	Time resolution	Calibration checkable on site
PC	Yes	Yes	No	Yes	No
LPI	Yes	Yes	No	Yes	No
MPS	Yes	No	No	No	No

Surface measurement instrumentation evaluation

nstrument	Practical limit	Repeatability	Reproduci-bility	Time resolution	Calibration checkable on site
C	unclear	No	No	Yes	No
as 2000	unclear	No	No	Yes	No

The analysis shows that only the gravimetric method completely fulfils the measurement requirements. The other mass-based methods may have the potential to fulfil the Table 1 requirements.

4 Conclusions

Influences of engine, fuel and measurement and sampling conditions on aerosol measurements

?? ***A new PM measurement metric which is sensitive to nucleation particles is potentially susceptible to large measurement artefacts and should therefore NOT be considered further.***

?? ***A combination of thermodenuder with size-unresolved total particle count cannot give valid data, because of size-dependent particle losses, and likely chemical species dependency on denuder efficiency.***

?? ***Because of the many parameters that must be kept constant and verified by regular calibration, in principal a thermodenuder is not suitable for regulatory measurements.***

?? ***A measurement metric based on 'solid' particle number is problematic because of the influence of aerosol dynamics on the measured value.***

?? ***Engine load and fuel dependent particle morphology changes can lead to unpredictable aerosol measurement effects. Examples include aerodynamic and mobility aerosol behaviour. Metrics influenced by this type of effect are neither reliable nor repeatable.***

- ***With a test procedure of practical relevance, no correlation between particle size and injection pressure of a modern, direct injection diesel engine was identified. As particles are formed in the gas phase, their size is not related to the droplet size in the diesel spray or to the injection pressure. Size resolved particle measurement gives no additional information and can therefore be replaced by mass measurements.***

Instrumentation related investigation results

- ***The particles generated by CAST show a stable average diameter (CMD), but a trend towards decreasing number concentration with time. It appears to be unsuitable as a calibration aid.***

- ***Measurements with the LQ1-DC in the particle range less than 70 nm are not plausible. The observed ageing effects make the general suitability of a corona charger questionable. Humidity and variation of the inner electrical insulation caused by deposited particles give a poor stability of the measurement signal. Additionally, no absolute particle surface calibration standard is available for system verification.***

- ***It is observed that the PAS 2000, in principal, is not capable of performing particle surface or mass measurements in exhaust gas from internal combustion engines with reliable and quantitative results. Additionally, no absolute calibration standard is available for system verification.***

- ***The reproducibility and comparability of ELPI measurements is limited unless a demanding maintenance/operation protocol is strictly observed. The ELPI is not suitable for performing regulatory measurements. Additionally, no absolute particle number calibration standard is available for system verification.***

- ***The SMPS does not fulfil basic requirements for quantitative or reproducible measurements. It is not capable of dynamic measurements. Additionally, no absolute particle number calibration standard is available for system verification*.**

- ***The CPC is very sensitive to nucleation effects. Due to the lack of size resolved information, these effects are not identifiable. The operation range is limited (variable high dilution needed) and the repeatability is low compared to state of the art PM mass methodology. Additionally, no absolute particle number calibration standard is available for system verification.***

- ***Measurement of elemental carbon by PASS or LII provides a sensitive and time-resolved measurement technique, which could in principle be absolutely calibrated by coulometry.***

- ***The measurement of primary particle diameter by LI^{2}SA does not offer notable size differences, therefore the usefulness of this parameter is questionable.***

?? ***The general applicability of the MasMo for regulatory engine exhaust measurements is questionable. In its current development status, it has low reproducibility compared to state of the art PM methodology. In addition, no absolute particle density calibration standard is available for system verification.***

?? ***The variation between several instruments (DMA+CPC, ELPI, DC) of the same type is very high even when they are simultaneously measuring the same particle source. Since a robust calibration method does not exist, these instruments are clearly not suitable for certification.***

?? ***For light duty vehicles the standard gravimetric procedure reveals the potential for particulate emission measurements also beyond Euro 4 regarding accuracy and the limit of detection. Optimization possibilities exist for further improvements.***

?? ***For heavy duty engines the PM measurement is possible down to a level of 0.01 g/kWh with a tolerable 10 % measurement error. Further improvements are feasible, if sulfur free fuel can be used. Taking into account this potential of the current type approval method, it should be retained for the future.***

The new measurement instruments can, as yet, only be used for the qualitative assessment of particulate number and size distribution as a relative comparison. They are still far from achieving the absolute quantitative measurement of these parameters. The measured particle number and size distribution can be manipulated with a careful choice of test conditions. Any new particulate measurement methodology should fulfil the same quality criteria as the current gravimetric method. Based on the above assessment, gravimetric measurement is considered to have the highest potential for future development. Other mass-based methods may have the potential to fulfil these requirements.

5 OICA contribution to PMP

Part 2 : Technical Annex (see: www.oica.net > Particulates)

5.1 Abgaszentrum der Automobilindustrie (ADA)

5.2 BMW

5.3 BOSCH

5.4 DaimlerChrysler

5.5 Ford

5.6 Volkswagen

5.7 Renault

5.8 JAMA