Working Paper No. WP-4
(GRPE Special informal meeting,
15 Sept. 2003, agenda item 2.)

Demonstration of feasibility of reproducible particle number measurements

1 Objective

The validation[1)] of the method for particle number measurement and the calibration of its components between tail pipe and output of the results at the particle counter, are feasible and lead to reproducible and reliable results.

2 Measurement of the distance-related particle number

2.1 Definition: Distance-related particle number

The quantity of measurement "distance-related particle number" *TPN* (total particle number) is the particle number per driven distance in a defined driving cycle. Particles shall be the solid components of the exhaust gas, which have a mobility diameter between 20 nm and 300 nm. The fluid components shall be neglected.

2.2 Example for Limiting Value

A proposed limit for TPN in a driving cycle (EURO 3: NEFZ) is $TPN_L = 10^{11}\ km^{-1}$. The measurement uncertainty must be considered in specifying the limiting value.

2.3 Basic requirements for the measurement method

- Measurement deviation: < 30 % (95 % confidence interval)
- Penetration of fluid particles neglected
- Discrimination of particle diameters below 20 nm and above 300 nm

2.4 Measurement method for the distance-related particle number

The measurement method for particles (Figure 1) has two parts. The first part is described in international standards (engine till CVS tunnel). But there exist no international regulation for the second part (sample extraction till particle counter).

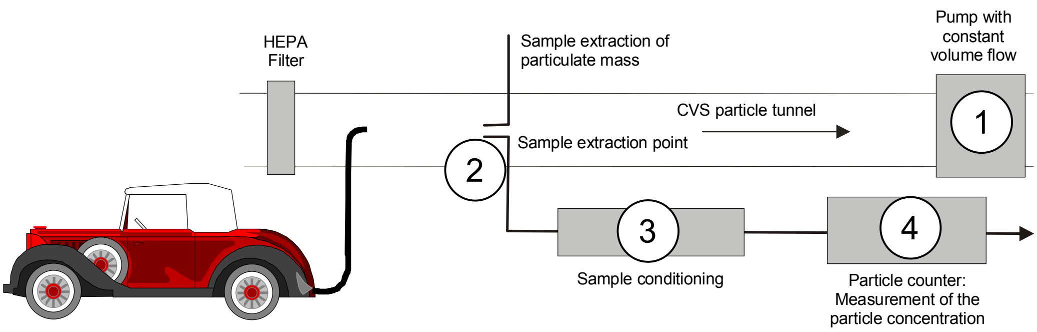


Figure 1 Schematic of the exhaust-gas particle measurement

1) The validation is an investigation of the pertinent influence quantities and their quantification. This knowledge establishes the requirements, the calibration procedures and it determines the measurement uncertainty.

1. Dilution of the entire exhaust-gas in constant volume sampler (CVS). CVS has a constant volume flow (dilution is variable). The sample volume V_{mix} comprises the entire particle emission during the driven distance *d*.
2. Sample extraction: Extraction with a specified probe at a predefined location in the CVS particle tunnel (particle losses expressed as penetration P_t).
3. Sample conditioning: Dilution and selection of the solid particles with a diameter between 20 nm and 300 nm (dilution factor *DF*, size discrimination with penetration P_g, selection of solid particles with penetration P_f).
4. Continuous measurement of the particle concentration C_p (number per unit volume at normal conditions) at a defined flow (without size discrimination) and with penetration P_m.

The distance related particle number *TPN* is calculated with above parameters using the formula:

$$TPN = \frac{V_{mix}}{d} \cdot DF \cdot \prod_j P_j \cdot \frac{1}{n} \cdot \sum_{i=1}^{n} C_p$$

2.5 Influence quantities

The influence quantities are parameters that impact the measurement of particle concentration. The impact can be direct (e.g. flow) or indirect (e.g. longer residence time and intensive coagulation). They cause either a systematic deviation or scatter in the measurement results. The influence quantities are closely linked to the measurement method employed. As far as possible, the systematic deviations are quantified and computationally corrected. If the deviations are not reproducible, then the scatter and therefore the measurement uncertainty increases.

2.6 Reference method or target specification?

The definition of the measurement quantity alone as in section 2.1 is inadequate to obtain consistent results. According to today's knowledge different instrument realisations lead to different results. Hence the particle number cannot (yet) be defined in a target specification.

However, it is possible to define a reference method. A reference method is a sufficiently described procedure, which is per definition correct [2)] . Such a particle number cannot be traceable directly to the base units (SI). Nevertheless, the different components shall be calibrated with traceable standards.

An investigation of the influence quantities must precede the description of the reference method. The sensitivity of the particle number to these quantities determines the requirements for the components (connection dimensions, temperatures, calibration quantities and periodicity etc.).

2) Examples of reference procedure: Exhaust gas measurement with CVS-Tunnel, HC measurement with FID-Detector or IR-Absorption, PM_{10}-Measurement

3 Reference method of measurement

3.1 Measurement set-up

The proposed method for the measurement of the particle concentration in a CVS tunnel comprises a hot dilution, an impactor (or cyclone) and a particle counter [3] (Figure 2).

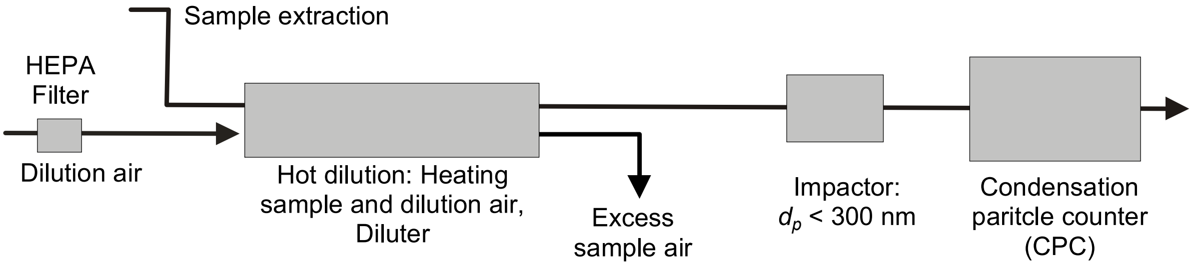


Figure 2 Schematic of the exhaust-gas particle measurement

3.2 Validation of the reference method

The reference method shall be tested for suitability, before it is employed:

- Variation in the mechanical dimensions (manufacturing tolerances or tolerances in the specification): sampling point, sample extraction, connections, etc.
- Abrasion, damage or soiling of mechanical parts
- Tolerances for thermostatic components (discrimination of fluid particles)
- Tolerances for sampling flow and dilution
- Shape of the particle size discrimination (nominal penetration curve $P_{nom}(d)$)

The Appendix lists the most important influence quantities and the possible requirements for the reference method.

3.3 Calibration of the reference procedure

The calibration of the reference method comprises the following steps:

- Calibrate the zero point (without particles)
- Calibrate the flows (diluter, counter)
- Calibrate the temperatures (hot dilution, CPC)
- Calibrate the particle number (and penetration) for the entire system (possibly using CVS) for at several particle sizes and concentrations using "dry" particles from a calibrated CAST (Example of the calibration in Figure 3).
- Determine the penetration of fluid particles (possibly with modified CAST[4]).

[3] VDI 3489 (December 1995): Messen von Partikeln, Methoden zur Charakterisierung von Prüfaerosolen: Kondensationskernzähler mit kontinuierlichem Durchfluss (Particle measurement, Methods to characterize test aerosols: Condensation particle counter with continuous flow)

[4] Fluid particles are added to the dilution air of the CAST (*see M. Fierz and H. Burtscher (2003) PMP report CH6)*

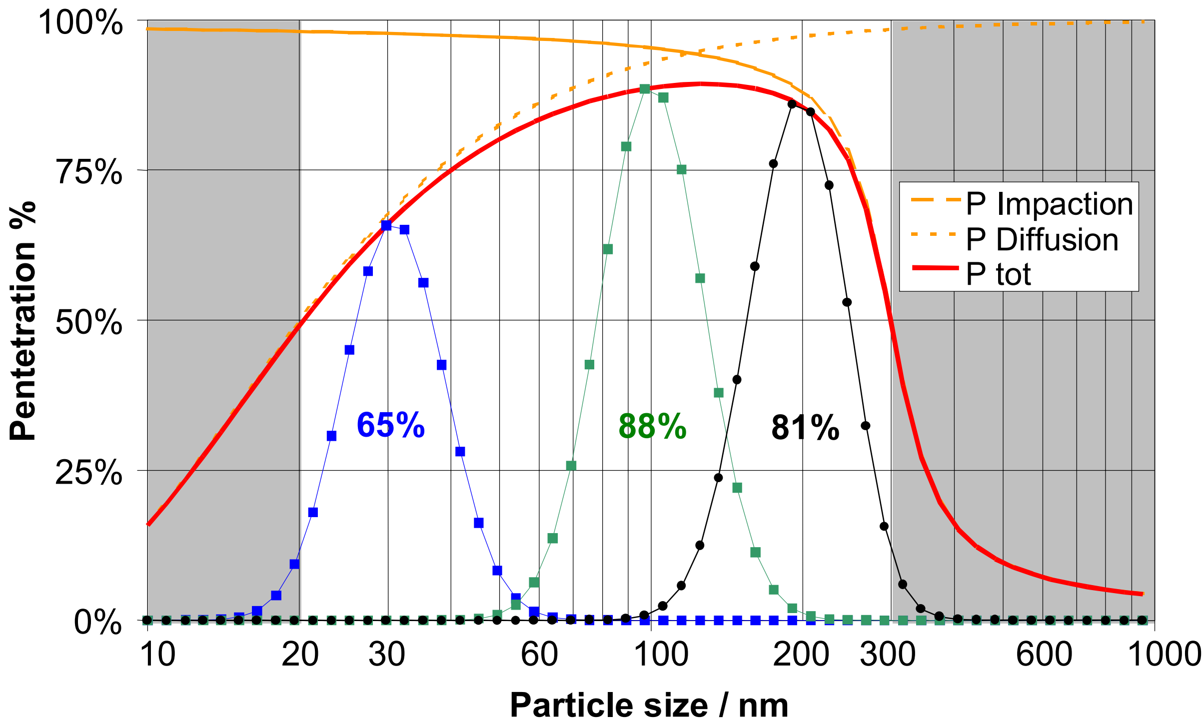


Figure 3 Theoretical example of a reference procedure: Size specific penetration from impaction (d_{50} = 300 nm) and diffusion (d_{50} = 20 nm) are together the total penetration P_{tot}. The calibration with 3 test aerosols from the CAST (σ_g = 1.6, d = 30 nm, 100 nm and 200 nm) spans the entire size range. The data are the penetration of the test aerosols.

3.4 Summary

The reproducible measurement of the distance-related particle number for diesel engines is

- feasible
  (introduced as a reference method of measurement)
- available
  (principal components are introduced to the market: hot diluter, impactor, particle counter)
- affordable
  (complement to the existing CVS)
- reliable
  (validation quantifies the uncertainties)
- ready in a little while
  (validation lasts about one year with PMP phase III)

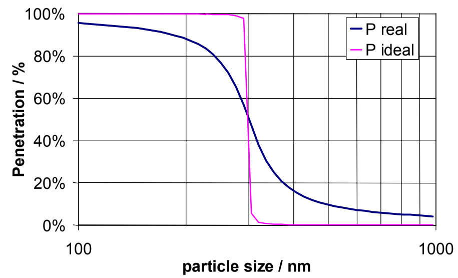
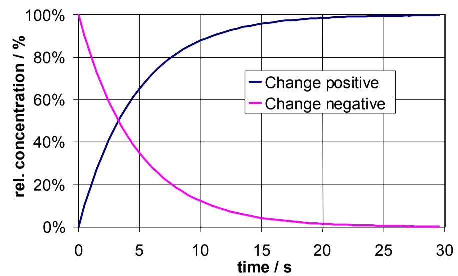
juerg.schlatter@metas.ch, 10.9.2003

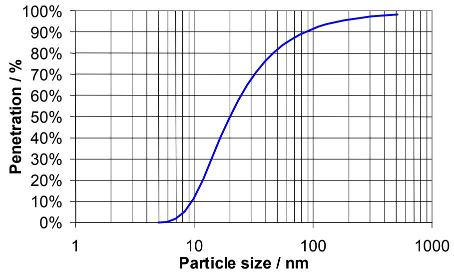
4 Appendix: Influence quantities and requirements for the reference method

The following table discusses the influence quantities of the reference method.

- The requirements are based on experience and will be adapted during validation. The final requirements are conditions for attaining the objective.
- Validation is a once only test of system suitability, assuming that any divergence will be detected through verification or calibration.
- Calibration is the (periodic) comparison of a quantity with traceable standards. Its periodicity must be adapted to the influence quantities.

Location	Cause	Effect/ Observation	R = Requirement; V = Validation; C = Calibration
All	Leaks	Falsifies the concentration	R: Leakage rate shall not impact measurement C: Calibrate the zero point before measurement
CVS Tunnel and diluter	Dilution air in CVS-tunnel and in hot dilution contains particles	Too high concentration	R: Without engine no particles: < 1 % TPM_L C: Calibrate the zero point before measurement
CVS Tunnel	Connecting pipes or CVS tunnel emit particles	Too high concentration	R: Without engine no particles: < 1 % TPM_L C: Calibrate the zero point before measurement
Extraction in CVS tunnel	Particle concentration not uni-form over the cross-section	Falsifies the concentration	R/V: Deviation of the probe position (3 directions): < 2 % of the mean value
Extraction in CVS tunnel	Sample extraction is not isoki-netic	Smaller flow ⇒ more concentration Greater flow ⇒ dilution	R/V: Difference of gas velocity between probe and tunnel < 20 %
Hot dilution	Size-specific deposition		R/V: Maximum volume: 0.1 L R/V: Through-flow rate: 1.00 ± 0.05 L/Min
Hot dilution	Condensation of exhaust gas components	New particles or non-separation of existing fluid particles	R/V: Dilution factor: 30 ... 1000 Setting (only) before the measurement R/V: Dilution gas and raw gas at dilution instant have a temperature of 195 ± 5 °C (during at least 0.5 s) R/V: Penetration of fluid particles < 1 % TPM_L C: Temperature calibration
Hot dilution	Dirt in the diluter	Modifies the dilution factor	R/V: Deviation of the dilution, depending on the size 20 nm ... 300 nm) during >10 tests, without maintenance: < 1 % *DF*

Location	Cause	Effect/ Observation	R = Requirement; V = Validation; C = Calibration
			R/V: Repeatability of diluter setting < 1 % *DF*
Impactor	Penetration (filtration characteristic) not ideal and depends on flow		R/V: d_{50} = 300 nm R/V: d_{80} > 250 nm and d_{20} < 400 nm V: Dependence of *P*(*d*) on flow C: Calibrate the flow V: Influence of pollution on *P(d)* C: Calibration *P*(*d*)
Particle counter	Flow rate depends on ambient pressure and temperature		R/V 900 ... 1050 mbar and 15 ... 30 °C, flow changes less than 1 % of nominal value C: Calibrate flow
Particle counter	Because of dead time, not all particles are detected in counting mode (at concentration > 1000 cm^{-3})		R/V: Error due to dead time < 1% at maximum concentration
Particle counter	Several particles arrive simultaneously, but are only counted single.		R/V: Warning at too high concentration
All	Short smoke peaks may not be measured, when response time is large. Differences in t_{90} cause deviations in particle number.		R/V: The response time of the entire system: 10 ... 20 s (t_{90} at step change of concentration in both directions)

Location	Cause	Effect/ Observation	R = Requirement; V = Validation; C = Calibration
All	Losses due to deposition in the entire system, and errors due to detection threshold of the particle counter. The separation of smaller particles is essential for comparative measurements.		R/V: d_{50} = 20 nm R/V: d_{80} < 50 nm and d_{20} > 10 nm V: Dependence on flow V: Influence of the pollution
All	Deposition of particles on the walls. Depends on geometry (bends, constrictions, enlargements, eddy formation), surface (roughness, charge), flow, residence time and temperature conditions.		R: Size-specific composite penetration within 5 % of nominal Penetration P_{nom} C: Calibrate the penetration at different particle sizes, with CAST, before each measurement R/V: Total volume does not exceed a maximum value R/V: Adequate radius of bends R/V: Surface is electrically conducting