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**Particle Emissions from Diesel Engines -
Measurement of Combustion Exhaust and
Occupational Exposure**

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Abstract: Recent research clearly indicates that it is the submicron fraction of the particulate pollution that is responsible for adverse health effects. The main source of submicron particles are combustion processes, first of all diesel engines. Means to measure the particles in combustion emissions, for occupational exposure, and in ambient air are therefore urgently needed. Whereas laboratory equipment is available, tools for field measurements are still missing. A setup which is useful for this purpose is presented. It consists of a conditioning part including precipitation of the coarse fraction, dilution and means for the removal of volatile material as well as sensors for particle concentration (condensation particle counter, CPC), particle surface (diffusion charger, DC) and a soot selective device (photoelectric aerosol sensor, PAS). All these sensors have a fast time response to also measure transients. Measurements on an engine test bench are used to demonstrate the tools. The carbon concentration correlates with the signal from PAS. The results also show that correct handling of the volatile fraction is very important to avoid severe artifacts, mainly due to particles arising from engines equipped with particle traps.

1. Introduction

The impact on public health of particles in diesel engine exhaust is a topic of increasing importance. Usually one has in mind diesel powered vehicles such as passenger cars, and most of all trucks.

However, a large number of engines is also used in off-road applications, e.g., for electrical power generation, marine and construction machines (BUWAL, 1996).

If these engines are operated in areas with reduced air exchange such as tunnels, serious problems may occur.

Table 1 shows typical concentrations for the most important pollutants in diesel exhaust and the corresponding Swiss limits for occupational exposure. To meet the regulations for the ambient air in such areas the exhaust gases have to be diluted by forced ventilation. It is evident that the required dilution is completely dominated by the particle emissions. Considering a typical exhaust volume of diesel engines of $6 \text{ m}^3/\text{kWh}$, a minimal dilution air volume of the order of $750'000 \text{ m}^3/\text{h}$ is required for a 100 kW-diesel engine to keep the particulate pollution of ambient air in tunnels below the legal limits. Beside technical problems this causes tremendous costs, and only a reduction of particle emissions can solve this problem. Progress in engine construction already has significantly reduced the emissions, however, this is by far not sufficient. Other techniques such as particle traps or fuel additives have to be applied.

Therefore, emissions from off road diesel engines and means for their reduction have been studied (Mayer et al., 1998) in the framework of a program called VERT (Verminderung der Emissionen von Realmaschinen im Tunnelbau).

mg/Nm ³	CO	NO _x	SO ₂	Particles
diesel engine emissions	1000	3000	350	250
MAK for occupational exposure	35	30	5	0.2
required dilution factor	28	100	70	1250

Table 1: Diesel engine emissions and limits (typical values for engines, used in construction sites)

Limits for occupational exposure are given for the elemental carbon (EC) concentration in Germany (TRGS 900, 1998) and the total carbon (TC) concentration in Switzerland (SUVA, 1997). The reference method for the determination of these concentrations is Coulometric analysis (VDI, 1995).

The emission limits, on the other hand, are up to now given by total mass (total particulate matter, TPM), determined by gravimetric analysis from filter samples, taken at a fixed temperature (52°C). At this temperature the volatile fraction is mainly in the condensed, i.e. the particulate phase.

The fact that different quantities are used for emission- and working place limits means that it is hardly possible to estimate working place limits from emission values, even if the dilution is well known.

This problem increases dramatically if particle traps are used. Good traps have an efficiency of the order of 99% for particles down to diameters of some ten nm. The EC fraction is therefore reduced by this factor. However, as the particle trap is close to the engine where the exhaust temperature is still high, much volatile material passes the particle trap in the gas phase and condenses later on, when the exhaust gas cools down. At the 52°C for the gravimetric sampling, this material appears as particles. Particle trap efficiencies in terms of gravimetric analysis are therefore much lower, 90% or less. The ratio TPM/EC before and after the trap is completely different and depends significantly on the temperature. The very high volatile fraction after the trap enhances the temperature dependence and makes a reliable measurement of TPM concentrations at working areas or ambient air measurements hardly possible when particle traps are used.

So far the problems of the particle mass measurement has been discussed from the point of view of reliability and reproducibility.

The other question of course is, which value is important from the point of view of health effects.

Recent research indicates that the submicron fraction which can penetrate into the alveolar region, is of crucial importance (Mohr et al., 1993, and Peters et al., 1997). This result leads to a reduction of the size limit from PM₁₀ to PM_{2.5}, but PM₁ is already in discussion.

Another approach is to change from mass to number- or surface concentration. This automatically gives more weight to the very small particles, which mainly determine the number concentration but are not very relevant for the total mass.

These considerations have a significant impact on techniques for particle measurement as methods are required to determine the submicron fraction and to get relevant information on the volatile fraction. So far it is not clear how regulation will develop in the future, whether particle mass in the small size range will be limited, or the number concentration, and how the problem with volatile material will be treated.

In any case, means for doing these measurements in the laboratory and monitoring instruments for field measurements will be required.

These instruments should

- be sensitive to the submicron fraction
- allow to distinguish between soot, volatile fraction and non-soot particles

Beside test bench measurements it is required to have means for periodical control of particle emissions of diesel engines and - if particle traps are used - of the trap characteristics. The measurement should be such that emission and occupational exposure / ambient air values can be compared, i.e. the same quantities should be measured in both cases, preferably with the same methods.

Whereas laboratory technologies are available to some extent, field measurements still pose considerable problems.

In the following we present a way to quantify particulate emissions in the field and in the laboratory.

2. Elements of a field measurement system

Figure 1 shows a schematic diagram of the proposed setup. In a first step the coarse fraction has to be removed. This is necessary mainly for working place measurements, where considerable amounts of dust may be present, while engine emissions usually do not contain many coarse particles.

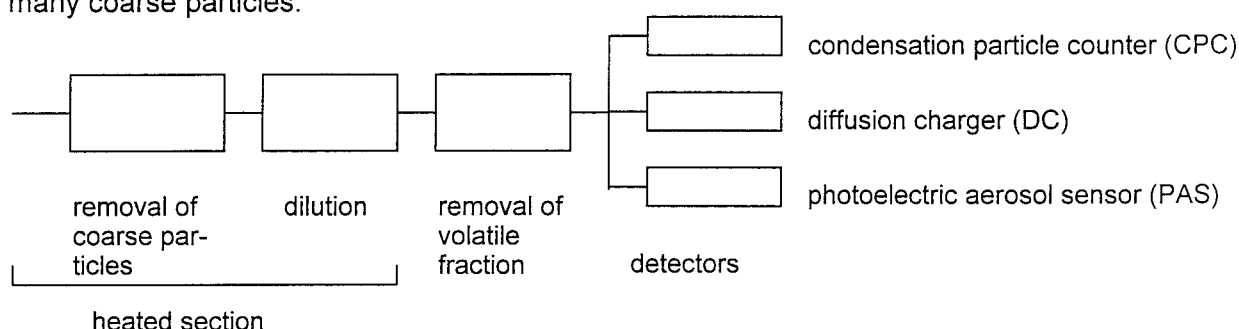


Fig. 1: Setup for field measurements

Dilution is required to obtain concentration levels which are adequate for the subsequent detectors, but also to prevent problems with condensation of vapors when the exhaust gas cools down. While the coarse particle precipitator (e.g. cyclone, impactor) and the dilution have therefore to be heated, the subsequent parts may be operated at ambient temperature. Note that the volatile fraction can be removed by a thermodesorber.

Useful detectors are

- the condensation particle counter (CPC) to measure the number concentration
- the unipolar diffusion charger (DC) followed by an aerosol electrometer which yields information on the surface concentration
- the photoelectric aerosol sensor (PAS) which allows to distinguish between soot and non soot fraction.

These components will now briefly be described.

2.1. Coarse particle precipitation

As already mentioned the precipitator for coarse particles is mainly required for working place applications where substantial dust concentrations may occur. Just for this purpose a heated system is not really required. Nevertheless, it is favorable to have a heatable precipitator which can be used also for emission applications (for example in combustion of solid fuels such as coal or wood) without running into problems with condensation of volatile material.

The coarse fraction can be precipitated readily by an impactor or a cyclone having a cut off diameter of a few μm . Cyclones can be operated also at high dust concentration, as they possess a large storage capacity for the precipitated material.

2.2. Dilution

In engine test bench applications dilution tunnels or mini tunnels (e.g. the AVL smart sampler) are used for complying the demands of the constant volume sampling (CVS) system of the European 13-Mode ECE R.49 or the ESC tests. These are very large and expensive devices which by no means can be used in field measurements. In other common dilution methods such as ejector type systems, the dilution factor depends on the pressure at the input, i.e. in the exhaust pipe of the engine to be measured. As this pressure usually changes during operation, this diluter is very problematic. A simple and effective way for dilution of submicron particles is described by Hüglin et al (1997), see figure 2. A rotating disk, containing a number of cavities transports a well defined amount of gas from the input channel, where the undiluted gas flows, to the output channel, where dilution gas flows. The dilution ratio is determined by the speed of rotation, number and volume of the cavities and the flow rate of the dilution air. However, it is independent of the flow rate of the undiluted gas. In particular, pulsation's from the engine are not transferred to the diluted side. This system allows stable dilution, adjustable from 1:10 to 1:10000. It is small and can be mounted close to the exhaust pipe, making long heated tubes unnecessary and avoiding or at least reducing artifacts such as coagulation in the sampling tube.

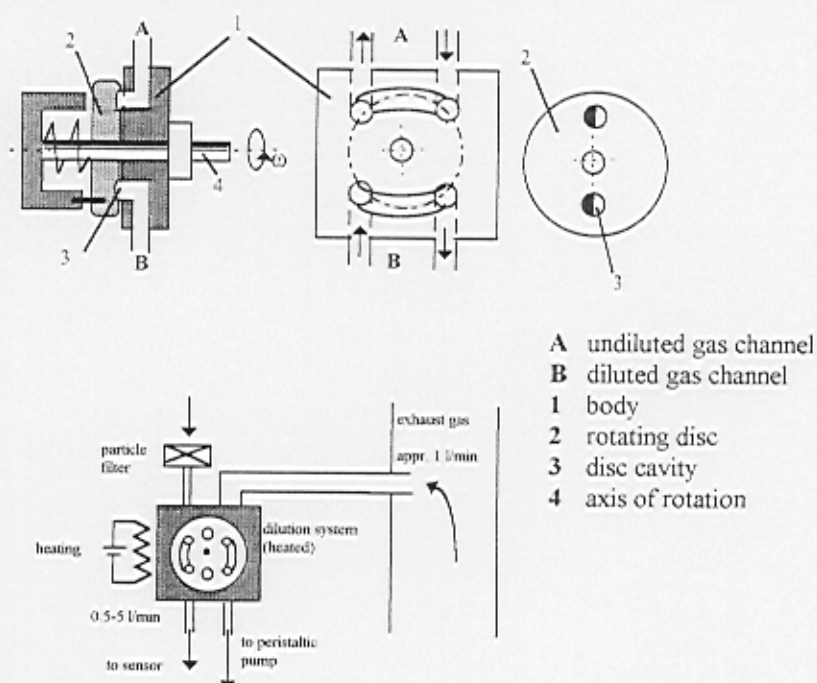


Fig. 2: Dilution system

Figure 2a and 2b show some characteristics of the adjustable dilution system for nanoparticle measurement. The dilution ratio is in an accurate linear relation to the speed of the rotation disk as shown in figure 2a. Figure 2 b illustrates particle size distribution measurements of 3 different aerosols with and without dilution verifying that the dilution ratio is independent of the particle size within the submicron range.

2.3. Thermodesorber

The thermodesorber (figure 3) to remove volatile material contains a heated section where the material is vaporized. It is followed by a water cooled section with walls consisting of activated charcoal. There the volatile material is absorbed to prevent reattachment to the particles as the temperature drops (for more details see Bertscher et al., 1995)

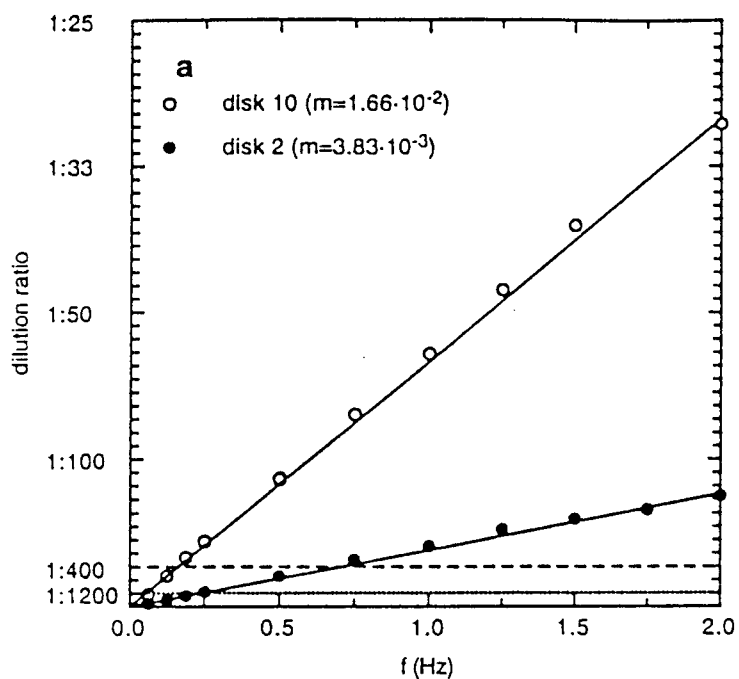


Fig. 2a: Dilution ratio as a function of the speed of rotation for a disk with 2 and 10 cavities

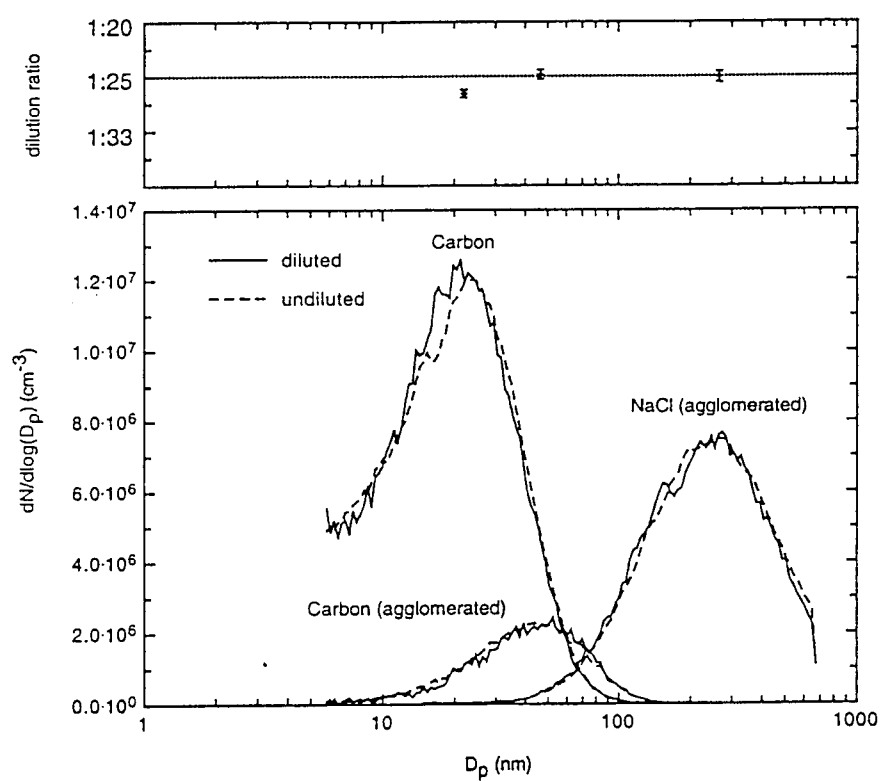


Fig. 2b: Comparison of SMPS size distribution for 3 aerosols with and without dilution.

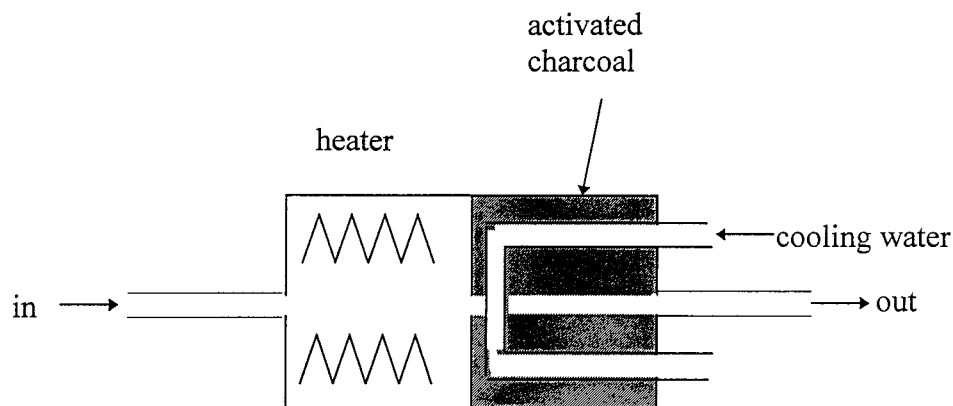


Fig 3: Thermodesorber

2.4. Detectors

2.4.1. Condensation particle counter (CPC)

Condensation particle counters (Stolzenburg et al., 1991) are suitable to determine the particle number concentration down to very small particles in the nanometer size range. For the measurements described below a TSI 3025 CPC has been used. This is a relatively expensive and delicate instrument which is of limited use in field measurements. For future applications it is planned to apply a simpler and more rugged device based on the same principle.

2.4.2. Diffusion charger (DC)

In the diffusion charger particles are charged by the attachment of ions of one polarity, created by an electrical corona discharge. In the subsequent aerosol electrometer the current, produced by the charged particles is measured. The attachment of ions to particles depends on the particle size, but not significantly on their composition (Adachi et al., 1985). In the free molecular limit the attachment coefficient is proportional to the square of the mobility diameter D , i.e. diffusion charging is a measure of the particle surface. In the continuum limit the attachment probability scales with D . As combustion particles mainly are small enough to be in the free molecular range, diffusion charging can be used to obtain surface related information. As for attachment of neutral atoms or molecules, the attachment coefficient is determined by the 'Fuchs-surface' (Fuchs, 1964). In the case of ions care has to be taken that the ion concentration is low enough to prevent errors due to the repelling Coulomb potential in multiple charging.

2.4.3. Photoelectric aerosol sensor (PAS)

Whereas diffusion charging is more or less material independent, photoelectric charging, i.e. the emission of electrons from the particle upon irradiation with UV light, is sensitive to chemical surface properties. In a number of studies it has been shown that this method responds to soot (Burtscher et al., 1994).

The charging probability in this case also scales with the particle surface area. Looking at the ratio of photoelectric charging to diffusion charging therefore eliminates geometry and concentration. What remains is chemical information, i.e. information on the chemical nature of the particle. This technique was used by Amman et al. (1992) to characterize volcanic aerosol. In the case of combustion particles this allows to distinguish between particles arising from different combustion processes. In figure 4 the PAS signal is plotted versus DC signal for different aerosols (Siegmann et al., 1998). If the slope is constant which actually is the case for most aerosols except cigarette smoke, the particles do not change during the experiment. The magnitude of the slope is characteristic for the source from which the particles were emitted. In this way, source attribution of particles is possible.

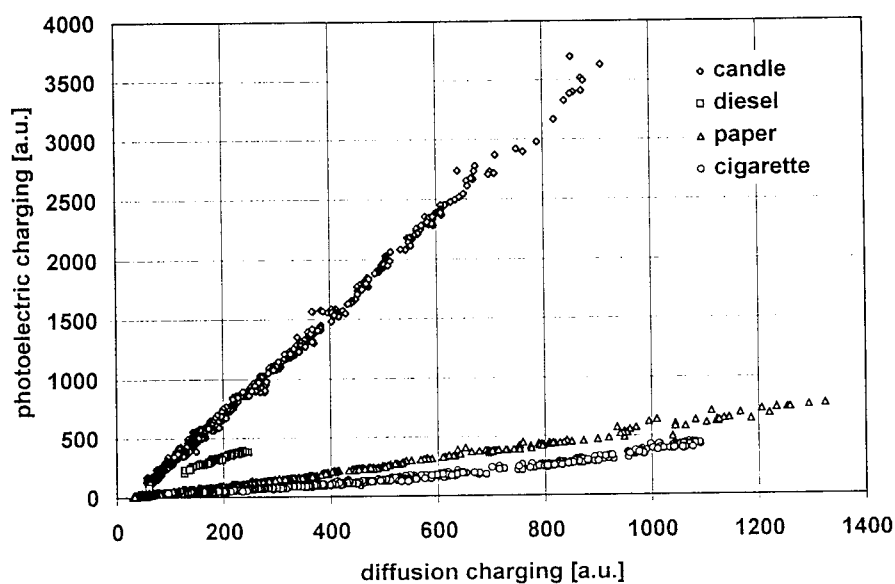


Fig. 4: PAS signal versus diffusion charging for different kinds of combustion

The PAS and DC sensors are shown in figure 4a. These two sensors are identical except for the charging. The charged particles are sampled in an electrically isolated particle filter. An electrometer amplifier converts the charge on the filter into a dc-voltage signal. The resolution and stability of the amplifier is better than 1 fA.

Photoelectric Charging (PC)

Diffusion Charging (DC)

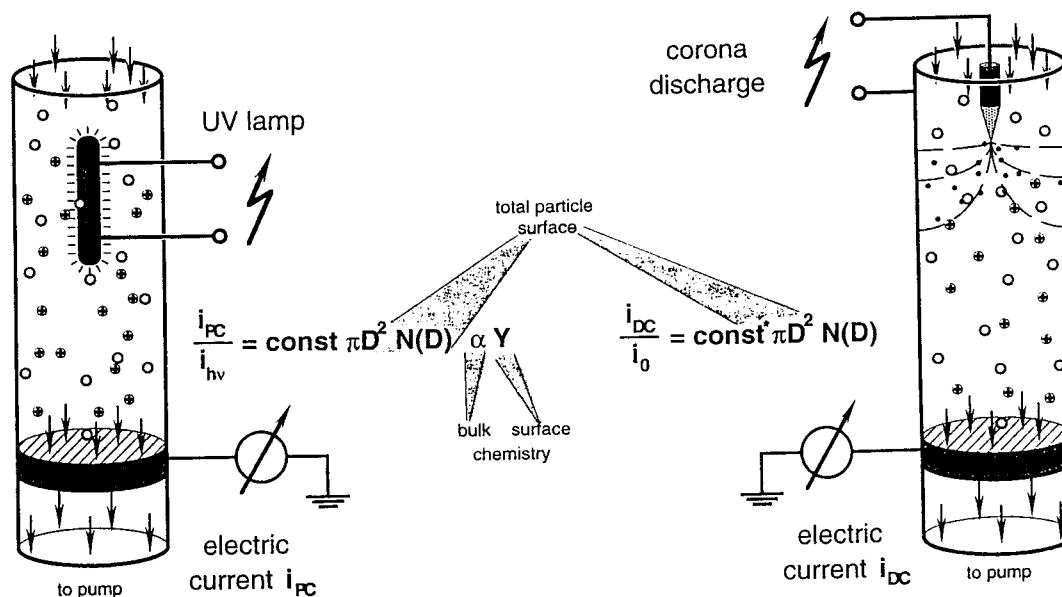


Fig. 4a: Principal diagram of PAS and DC sensor

i_{PC} : photoelectric charging current
 i_{DC} : diffusion charging current
 D : mobility diameter

i_{hv} : UV lamp current
 i_0 : corona discharge current

3. Test bench results

The system described above has been compared in a large number of measurements on a engine test bench with

- Scanning Mobility Particle Sizer (SMPS)-measurements, which yield the particle size distribution (Wang et al., 1990)
- aethalometer measurements. The aethalometer measures optical absorption and thus is sensitive to the black carbon concentration (Hansen et al. 1984)
- gravimetric and coulometric analysis

The test bench is operated with a dilution tunnel (AVL smart sampler) downstream and an adjustable dilution unit as described above upstream the particle trap. This setup makes possible the simultaneous measurement on both sides of the trap. The diesel engine is a 105 kW Liebherr TDI engine type 914 T for construction machines, and the particle trap is a SHW sinter-metal filter. Both PAS are operated simultaneously while the SMPS measurements are done alternately up and downstream the trap. Fig. 5 shows the setup used for these measurements

Fig. 6 shows a typical example of a size distribution before and after a sintermetal particle trap, determined by a SMPS-measurement and the trap penetration. This shows that the particle trap penetration is very low and does not significantly vary with particle size. Also plotted in the same graph is the ratio of the PAS-signal upstream and downstream the particle trap. This integral value for the particle trap penetration agrees with the SMPS-result. The engine was operated in steady state conditions at 1400 rpm and 50 % of the rated torque. Similar results for the particle trap penetration within ± 20 % variation are obtained at 3 additional operating conditions 1400 rpm/100% torque, 2000 rpm/ 50 % torque and 2000 rpm/ 100 % torque.

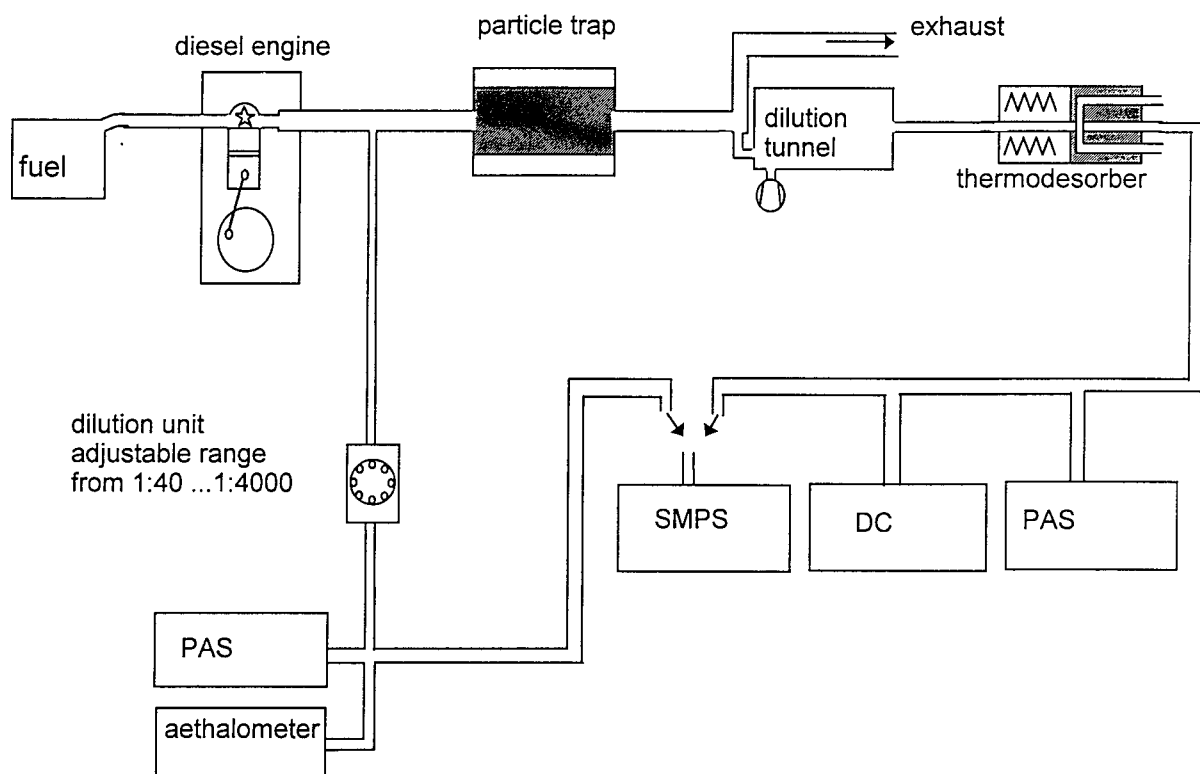


Fig. 5: Setup of the test bench measurements

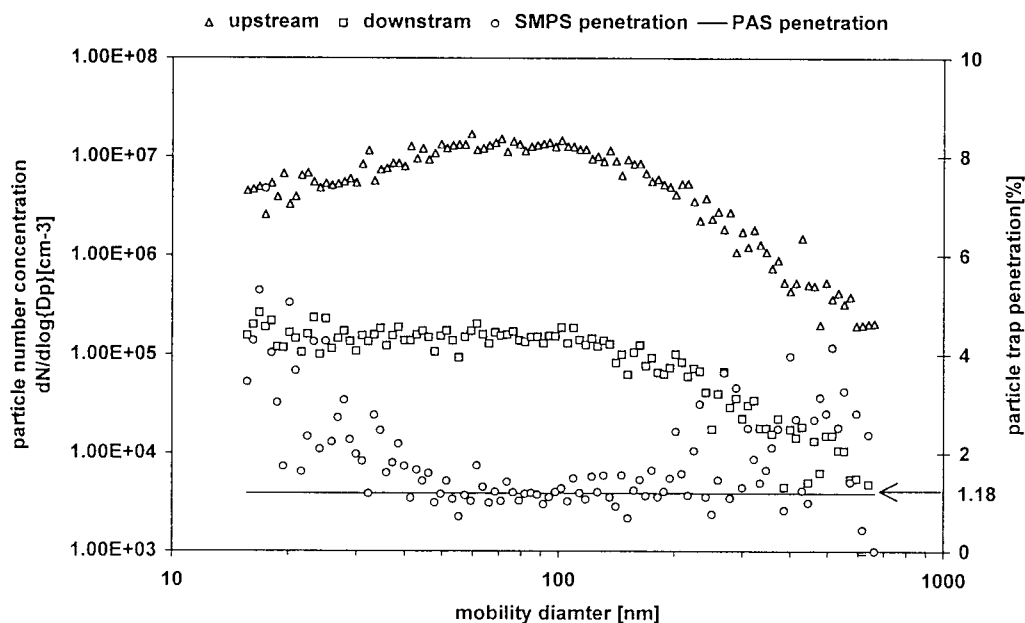


Fig. 6: Size distribution of particles in the emissions of a heavy duty diesel engine up- and downstream the particle trap. The trap penetration is also given.

The good agreement between the 2 different observations demonstrates that the PAS provides an integral information on the penetration characteristics of particulate trap in the sub-micron size range.

In certain operation conditions (high temperature in the trap) high concentrations of very small particles are found after the trap, in some cases much higher than upstream. These particles are mostly ultrafine H_2SO_4 droplets formed from SO_3 in the cooling down phase downstream the trap (Gairing et al. 1994). SO_3 is generated in most traps and catalysts from SO_2 when the temperature in the trap is above approximately 460°C . Additionally these particles can be caused by condensation of volatile material, passing the trap in the gas phase and then condensing as the exhaust gas cools down. That these particles are volatile can easily be proved by varying the temperature in the thermodesorber, as shown in figure 7. The engine is driven on full load whereby a temperature of $> 500^\circ\text{C}$ is reached in the trap. The number concentration of the volatile particles is more than 2 decades higher than that of the soot particles which penetrate the trap. At higher temperatures in the desorber, the volatile species evaporate first and are retained afterwards in the desorber. The thermodesorber is therefore a very important tool to distinguish between nonvolatile particles from the engine and volatile particles formed in the trap as well as to evaluate true penetration characteristics of traps.

Within the VERT project it has been found that in all tested diesel particle traps H_2SO_4 droplets with mobility diameter up to 50 nm and number concentration up to $10^8/\text{cm}^3$ were formed when the temperature in the trap exceeded $\approx 460^\circ\text{C}$.

The volatile material may also be distinguished from the primary C-particles by using the ratio of PAS and DC signals. Figure 7a shows the DC and PAS signals for the measurement at the same operating conditions as shown in figure 7. Whereas the PAS signal is only sensitive to the carbonaceous solid part but not to the volatile material, the DC measures the total surface concentration. DC is thus drastically increasing with decreasing desorber temperature whereas PAS even decreases. The PAS signal decrease is due to the very high surface concentration of the volatile aerosols. Electrons emitted from the soot particles are captured by the volatile aerosol instead of being removed thus providing a negative contribution

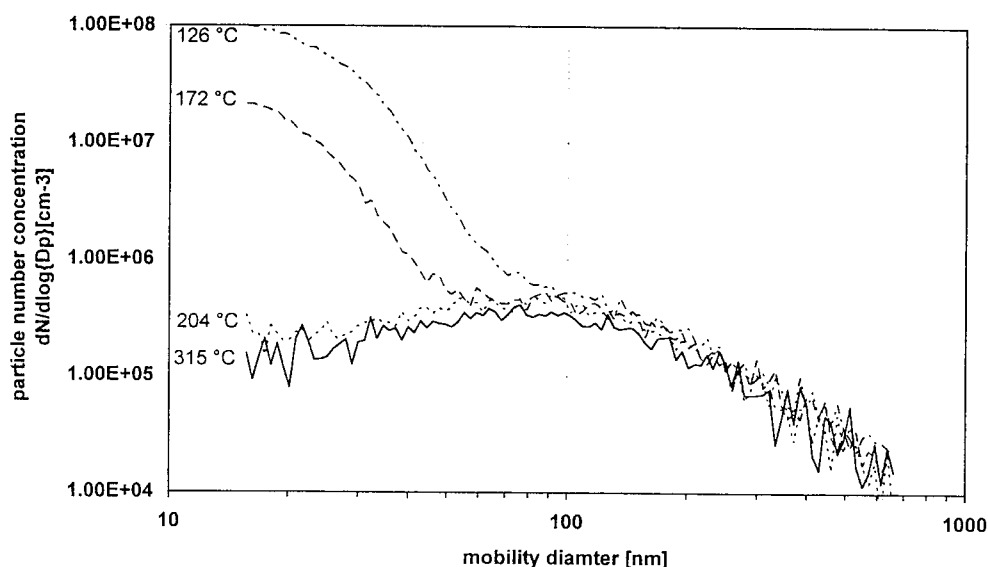


Fig. 7: Size distribution downstream the particle trap at different temperatures of the thermodesorber.

to the positive charge of the solid particles. This error is detected by increasing the dilution ratio or even better, by separating the volatile material in a thermodesorber. Therefore, using the 2 sensors with diluter and thermodesorber provides new information about submicron aerosols, specifically their chemical characteristics in exhaust systems with aftertreatment devices.

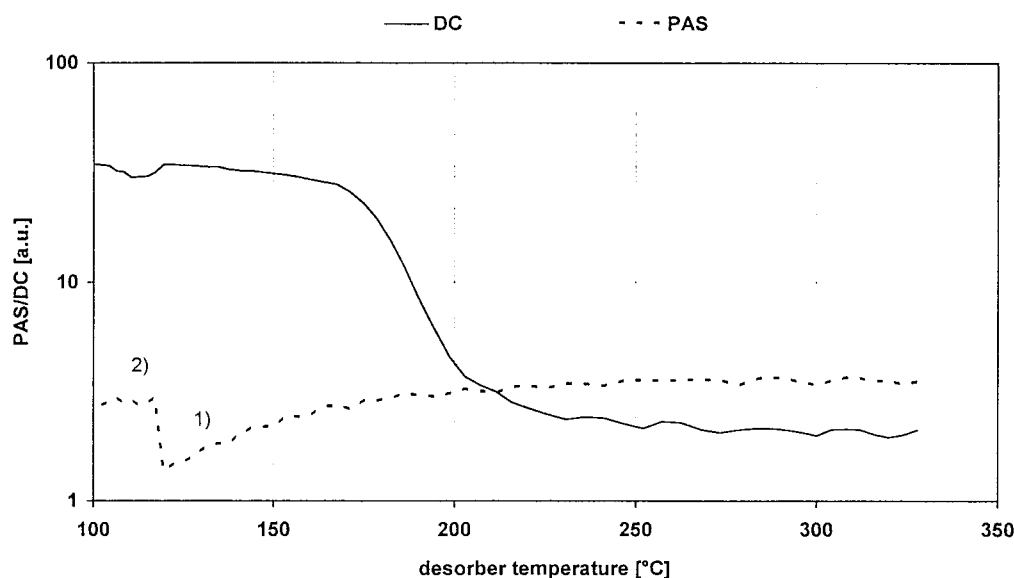


Fig. 7a: DC and PAS signal downstream a particulate trap at different temperatures of the thermodesorber. 1): dilution ratio = 1:8, 2): dilution ratio = 1:350

Figure 7b shows another application of PAS and DC for characterizing submicron particles from diesel engines. Within the VERT 7 program (Mayer et.al., 1998) it has been found that fuel with additive can create additional nanoparticles in diesel engine exhaust depending on the additive dosing rate and engine operation conditions. These particles have been identified to be oxide of the additive material. The photoelectric yield of the surface of these particles is less than 10 % compared to the one of the carbonaceous soot particles. Figure 7b shows size distributions of particles from the diesel engine (without trap) with and without additive in the fuel. The additive induces a significant decrease of the soot particles above

50 nm but ultra-fine oxide particles below 50 nm are formed in large number concentrations. Since DC measures all submicron particles but PAS only the carbonaceous soot fraction, the ratio between DC and PAS is markedly different for the 2 measurements as shown in the diagram on the right. Again comparison of PAS and DC provides a fingerprint of the chemical composition of the submicron particles.

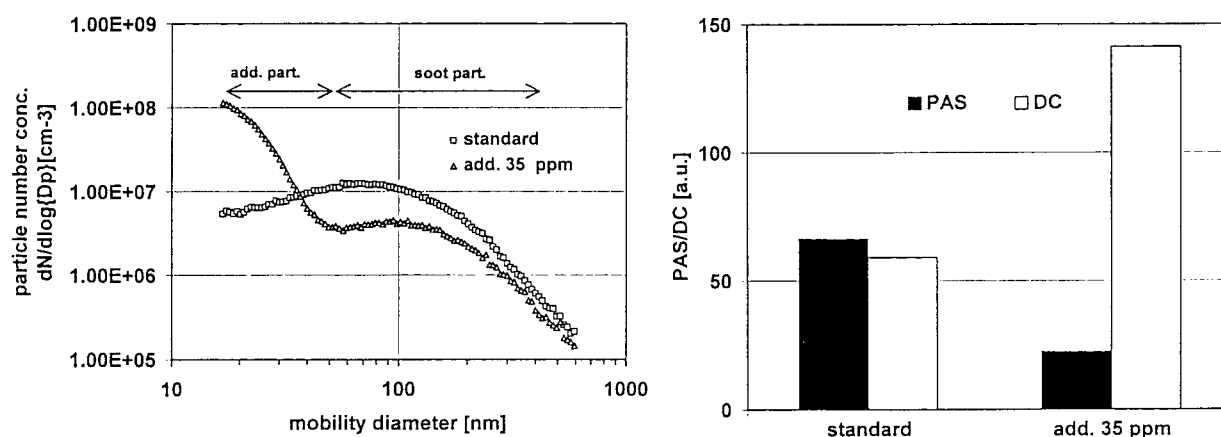


Fig. 7b: Size distributions and PAS/DC-measurements of submicron particulate diesel emissions with and without additive in the fuel.

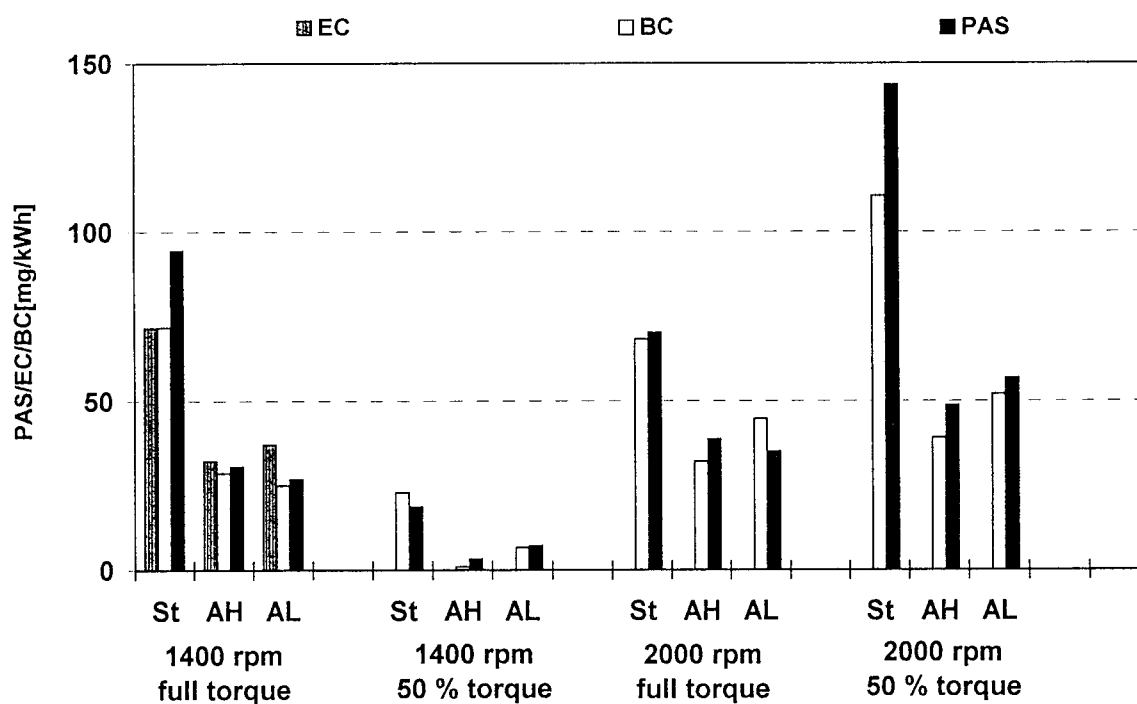


Fig. 8 Comparison of EC and BC concentration and signal of the PAS for various points of operation and fuels (St = standard fuel, AH = additive with rated dosing, AL = additive with 50% of rated dosing).

The next plot (Fig. 8) shows a comparison between PAS signal, EC concentration determined by coulometric analysis, and black carbon concentration (BC) measured by the aethalometer for various steady state operation conditions using standard fuel and fuel containing two types of additives. First, this shows that the benefit of the additives for particle reduction is quite limited. Much effort has been taken to find additives which efficiently suppress soot emissions, however, up to now the main incentive to use additives is to assist regeneration of particle traps. Secondly, the good correlation between PAS-signal and EC as well as BC concentration indicates that the PAS monitors the carbonaceous particulate fraction in diesel exhaust. This is valuable because it is particulate EC that is limited for occupational exposure.

Some of the legal emission tests on combustion engine emissions are performed in transient operating conditions, e.g., drive cycles on roller dynamometers. For gravimetric or coulometric analysis, filter samples are taken over the whole drive cycle. The short time response of the PAS allows dynamic measurements, yielding detailed information in which part of the cycle emissions are high or low. If the analyzer of the SMPS is set to one particle size, the subsequent CPC can also track the time dependence of the particle concentration.

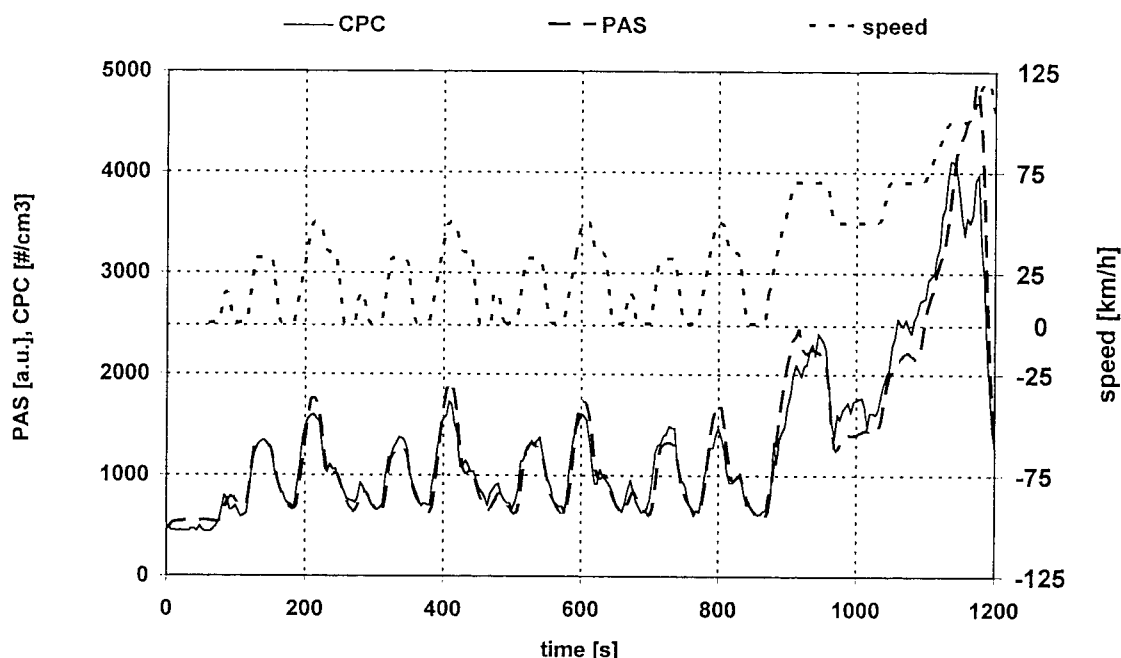


Fig. 9: Transient NEDC driving cycle, measured by the PAS, and the particle concentration at the outlet of a mobility analyzer set to 100 nm

Fig. 9 illustrates the PAS signal in comparison with the number concentration of 100 nm soot particles of a TDI diesel passenger car in an ECE-EUDC driving cycle on a roller dynamometer. SMPS measurements show that the mean mobility diameter of the particles is approximately 100 nm independent of the load condition. An excellent correlation is observed between the number concentration at 100 nm diameter and the signal of the PAS. This shows that photoelectric charging is tightly related to the concentration of particles in the submicron range where most of the soot particles are found.

Measurements on a test bench or a roller dynamometer are very expensive and therefore cannot be used for periodical tests of engine emissions or particle trap characteristics. A simple technique uses free acceleration (VRV, 1992). Even if this method yields much less detailed information, it may be sufficient for field tests, provided that an adequate measurement technique exists. The high time resolution of the PAS allows to perform such measurements in the field. Figure 9a illustrates dynamic measurements of free acceleration runs with PAS in comparison with a CPC at the output of a mobility analyzer set to 100 nm.

Trap penetrations are evaluated by the ratios of the PAS and CPC peak areas up- and downstream the trap from the acceleration runs. 2 PAS were measuring simultaneously in all runs whereas the CPC was connected downstream the trap for run 1 - 3 and upstream the trap for run 4 - 6. Table 2 shows the penetrations evaluated from these measurements.

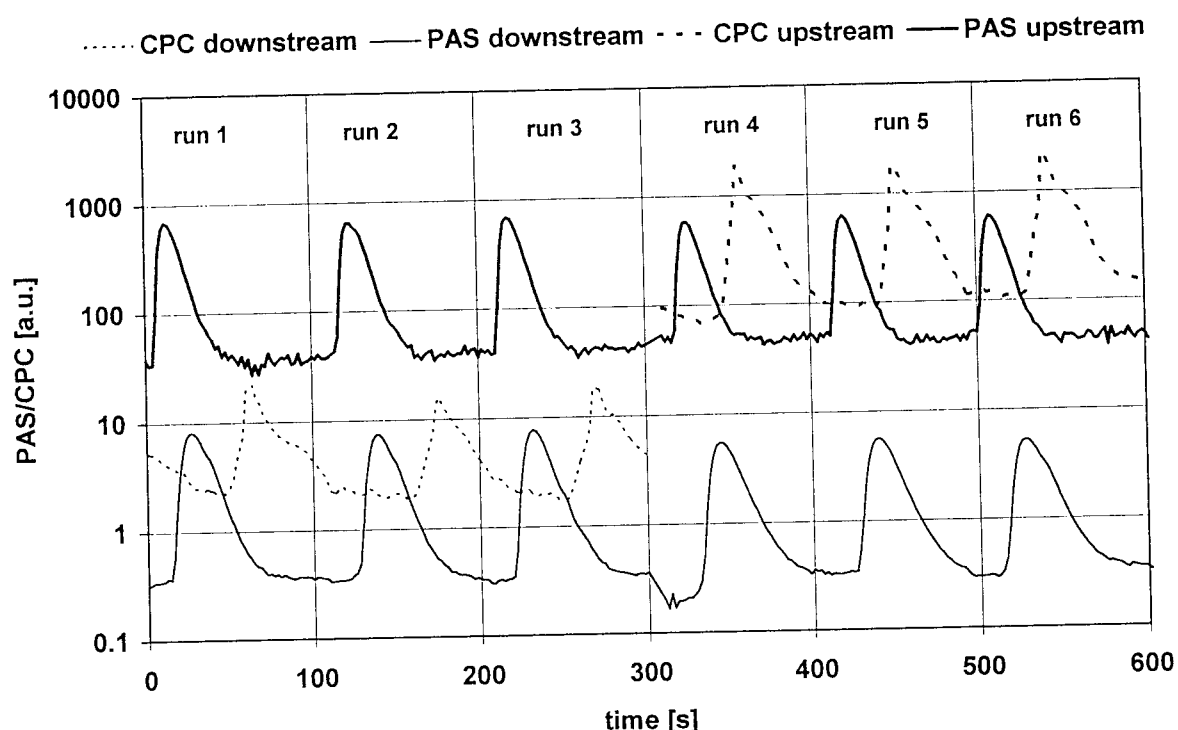


Fig. 9a: Free acceleration runs measured up- and downstream a particle trap by PAS and CPC at the outlet of a mobility analyzer set to 100 nm

	penetration [%]					
	run 1	run 4	run 2	run 5	run 3	run 6
PAS	1.62	1.37	1.30	1.34	1.39	1.38
CPC	1.30		0.90		0.93	

Table 2: Penetration of particle traps evaluated from free acceleration measurements

4. Conclusions

The test bench measurements with heavy duty construction diesel engines demonstrate that the emission of particles and their concentrations as well as the penetration of particle traps may be evaluated by fast and simple techniques.

The particle concentrations for occupational exposure can be determined in the same way.

The PAS (photoelectric aerosol sensor) responds to the carbon particles emitted by diesel engines.

The new dilution unit replaces the bulky dilution tunnels and is adjustable to a wide range of dilutions. It furthermore is portable, easy to use, and easy to maintain.

The removal of the volatile particles, for instance in a thermodesorber, is required to obtain results that are reproducible and can be interpreted.

PAS and DC provide relevant information on the chemistry, source, and the surface concentration.

As size classification of Diesel particles up to now has been the most complicated step of the particulate measurements, the presented system and procedure demonstrate great advantage.

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