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Field measurement of particle size and number concentration with the Diffusion Size Classifier (DiSC)

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ABSTRACT

The Diffusion Size Classifier (DiSC) is a new instrument to measure number concentration and average diameter of nanometer sized particles in the size range 10 - 200nm. It is small, easily portable and battery operated and therefore well suited for field measurements. The measurement range is suitable for ambient air concentrations (1000 – 500000 particles/cm^3); together with a diluter it can be used for emission measurements. The number concentrations measured with DiSC agree well with those measured with a condensation particle counter. The response time is short enough to measure transient engine operation. The DiSC is therefore a useful instrument for number concentration measurements in non-laboratory settings.

INTRODUCTION

Engine optimization and in particular the use of particle traps reduces the particle emissions of modern diesel engines significantly. A consequence of this reduction is that the emission measurement techniques used today are no longer adequate. The gravimetric method used for type approval tests is at the limit of its sensitivity. Besides, the condensation of volatile material in the measurement system can disturb the measurement (1). Opacimeters and smoke number meters used for field testing are by far not sensitive enough to measure downstream of a good particle filter. In addition, there are many discussions on whether total mass is a sufficient metric. The European Union recently decided to add a number based limit to the existing mass limits (2). A technique to measure number concentrations of solid particles has been developed in the framework of the Particle measurement program (PMP) (3), and it has already been extensively tested (4). It yields reliable results when used for type approval tests in the laboratory. However, the procedure is based on a dilution tunnel and therefore not portable and too expensive for field applications. So far no satisfactory number concentration measurement method for field testing exists.

Unipolar diffusion chargers are simple instruments, which have the potential to be sensitive, robust and portable. They are based on the electrical charging of particles by a corona discharge. If the current of charged particles is measured directly by an aerosol electrometer, this yields information related to the attachment cross section of the particles (5). The combination of a diffusion charger with a diffusion battery with several stages can be used to determine the particle size distribution roughly (6). As will be shown below, a diffusion charger with only one diffusion stage is sufficient to determine the particle number concentration, and, in combination with a backup filter as aerosol electrometer, also the average diameter. We developed a device based on this principle, the diffusion size classifier (DiSC).

PRINCIPLE OF OPERATION

Figure 1 shows the setup of the DiSC: First the particles are charged in a diffusion charger. Ions are produced by a corona wire, which is held at a positive high voltage.

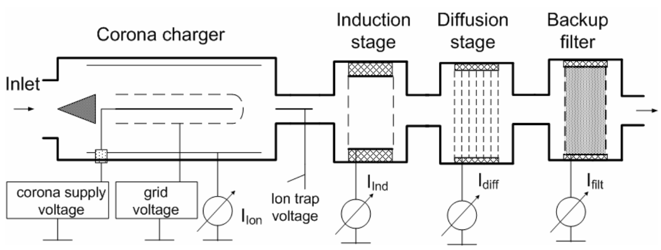

Figure 1: Components of the diffusion size classifier

A wire mesh separates the corona area from the area where the particle flow passes by to avoid particle losses due to electrostatic precipitation. A part of the ions produced by the corona penetrates the grid and may then attach to the particles. The ion concentration is kept constant by controlling the corona voltage so that the current I_{ion} produced by ions arriving at the electrode on the opposite side is constant. The probability of an ion attachment to a particle is given by the attachment coefficient K. The attachment coefficient for molecules is proportional to the inverse of the mobility b of the particles (7). By assuming that the attachment coefficient for ions and neutral molecules are equal (which is a strong simplification) the average charge $\overline{q}$ that a particle carries will be proportional to K, and thus b^{-1}.

$$\overline{q} \propto K \propto b^{-1} \qquad \text{Eq. 1}$$

Most of the ions do not attach to the particles in the charger. These free ions are removed in a subsequent ion trap by a small electric field. After leaving the charger and ion trap, the particles pass through an induction measurement stage (see later) and enter into a diffusion stage, which consists of several stainless steel grids. This stage is mounted electrically insulated and is connected to a current amplifier. The particles that are precipitated in the diffusion stage produce a small electrical current, which is measured with the current amplifier. This current I_{diff} depends on the particle number concentration N, the average particle charge, and the deposition probability by diffusion. If only a small fraction of the particles is deposited by diffusion, then the deposition is proportional to the diffusion coefficient, and thus:

$$I_{diff} \propto D \cdot \overline{q} \cdot N \qquad \text{Eq. 2}$$

The diffusion coefficient D is determined by the Einstein relation D = b kT and thus

$$D \propto K^{-1} \qquad \text{Eq. 3}$$

Replacing D in equation 2 by K^{-1} (according to equation 3) and $\overline{q}$ by K (according to equation 1) the diffusion current I_{diff} becomes

$$I_{diff} \propto K^{-1} \cdot K \cdot N \propto N \qquad \text{Eq. 4}$$

Average charge and diffusion coefficient cancel, and as a result the measured current is directly proportional to the particle number concentration.

As already mentioned, this calculation is strongly simplified and only intended to show the principle of operation. In reality the mean free path of neutral molecules and ions differs significantly which leads to a different particle size for the transition between free molecular range and continuum range. Nevertheless, the postulated relation holds fairly well, if the diffusion battery is designed such that only small fraction of particles is precipitated, as the experimental result in figure 2 shows.

The corresponding charging efficiency (average charge per particle as function of particle size) is shown in figure 3.

Particles penetrating the diffusion stage are captured in the following absolute filter, which is also connected to an electrometer. This amplifier measures the current I_{filt}. The current I_{filt} is related to larger particles, I_{diff} to smaller ones. The ratio of the two currents is therefore a measure of the average particle size. The ratio of filter current I_{filt} to the diffusion current I_{diff} can be calculated as function of the mean particle diameter. The result is shown in Figure 4, both for monodisperse particles as well as for lognormal size distributions with different widths.

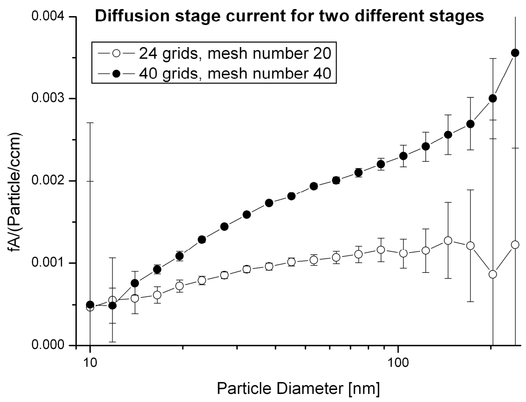

Figure 2: Current per particles as function of particle size. If a small number of diffusion grids is used, the size dependence is only weak. When more grids are used, the smaller particles are completely deposited in the diffusion stage, and so the measured signal is proportional to $\overline{q}$, and no longer independent of particle size.

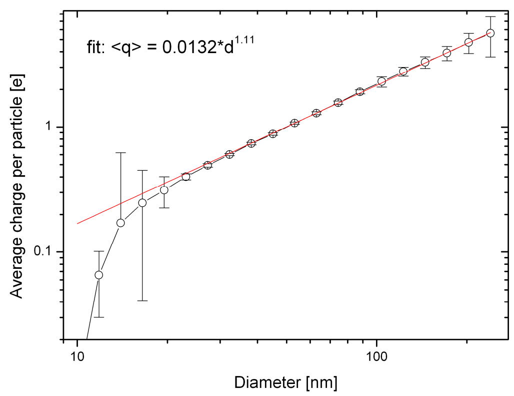

Figure 3: Average number of elementary charges per particle after the diffusion charger as function of particle size (dots). The solid red line is a power law fit ($q \propto d^x$).

It is clear from figure 4 that the calibration of the device depends on the shape of the particle size distribution. However, at least for engine emissions, the size distribution is lognormal and sigma is fairly constant (8); the error will therefore be small.

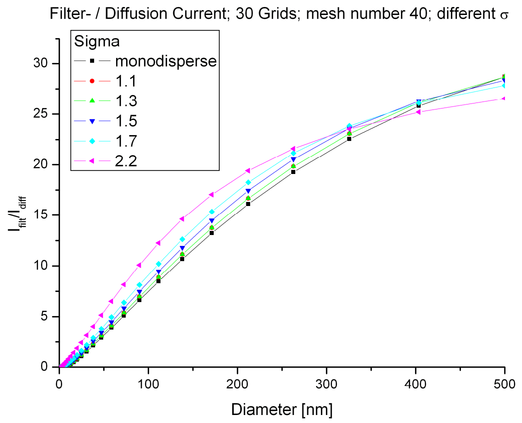

Figure 4: Calculated ratio of filter current to diffusion current (I_{filt}/I_{diff}) assuming a lognormal particle size distribution. Parameter is the geometric standard deviation σ.

Charged particles approaching the diffusion stage induce a current in the stage. If they are not precipitated in the stage the same current but with opposite polarity will be induced when they leave the diffusion stage. This induced current is superimposed to the current produced by precipitated particles, and thus produces a measurement artifact. The already mentioned induction stage is added to compensate for this artifact: The induction stage is a Faraday cage through which the aerosol passes. No particles are precipitated in the induction stage, but the induced current I_{ind} will be the same as in the diffusion stage (except for a small time shift). The “real” diffusion current is thus obtained by subtracting the induced current from the measured diffusion current.

We have implemented the system as described above in a small and portable device shown in Figure 5. It is battery operated; up to 12 hours of operation are possible on one battery charge. Both USB and Bluetooth interfaces are available. The DiSC can be operated from either a PC or a PDA. Standalone operation with internal data storage on an MMC card is possible too. As mentioned above, the suggestion resulting from the PMP program requires the determination of the particle number concentration for solid particles only. Volatile nucleation particles have to be removed from the gas stream before the measurement. To achieve this, a tube heatable up to 200°C can optionally be placed between the DiSC inlet and the diffusion charger. Nucleation particles, which may have formed, will evaporate there. If the dilution prior to the instrument is sufficiently high, no renucleation will occur when the gas cools down again. The battery operation time is reduced to about 6 hours by the additional power consumption of the evaporation tube.

The time response of the instrument is in the order of one second, the particle size range about 10 to 200 nm. The system is suitable for ambient concentrations (1000 - 500000 particles/cm^3). An external dilution unit is required when using it for emission measurements. A compact dilution system controlled from the DiSC is under development. It is based on the rotating disk diluter (9).

Figure 5: DiSC and PDA, connected by a wireless Bluetooth link

CALIBRATION

DiSC is calibrated with monodisperse aerosol. The ratio I_{diff}/I_{filt} is measured in a laboratory setup for particle diameters between 10 and 240 nm. Additionally, a CPC (TSI 3025) is operated in parallel to the DiSC. As a result, both I_{filt}/I_{diff} and the average charge per particle are measured for the particle size range from 10 to 240 nm. Figure 6 shows the resulting current ratio of such a calibration with Ammonium sulfate aerosol particles.

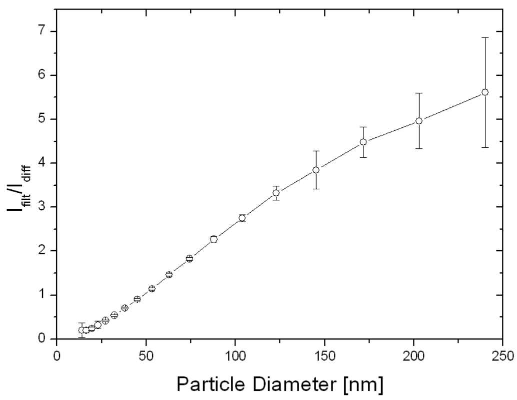

Figure 6: Measured ratio I_{filt}/I_{diff} for Ammonium sulfate particles. The line is a guide to the eye. Error bars increase towards the lower and upper end of the calibration size range because there are fewer monodisperse particles there.

From the measured I_{filt}/I_{diff} as function of the particle size, a polynomial regression can be performed, which relates I_{filt}/I_{diff} to the particle diameter as

$$d = \sum_{i=0}^{n} a_i \cdot \left(\frac{I_{filt}}{I_{diff}} \right)^i \qquad \text{Eq. 5}$$

In practice, a simple linear regression (n = 1) is sufficient for particles smaller than approximately 150nm, since the relationship between I_{filt}/I_{diff} and the particle diameter is nearly linear in this size range, as can be seen in Figure 6. A higher order fit is necessary for larger particles. As soon as the particle diameter is determined by use of Eq. 5, the particle number can be calculated with the power law fit for the particle charge:

$$N = c \cdot \frac{I_{filt} + I_{diff}}{d^x} \qquad \text{Eq. 6}$$

The data inversion procedure for both number a concentration and particle diameter is very simple and can thus be performed online even with only very small computing resources.

Every instrument has to be calibrated individually, because the feedback resistors in the electrometer circuits have large tolerances.

RESULTS

Figure 7 shows the number concentration measured during a transient driving cycle. A rotating disk diluter is used for dilution. The particle concentration is measured by a condensation particle counter (CPC, TSI 3010) and the DiSC. As the graph shows, the two systems give very similar results. It also shows that the time response of the DiSC is fast enough to follow the transients occurring during a driving cycle.

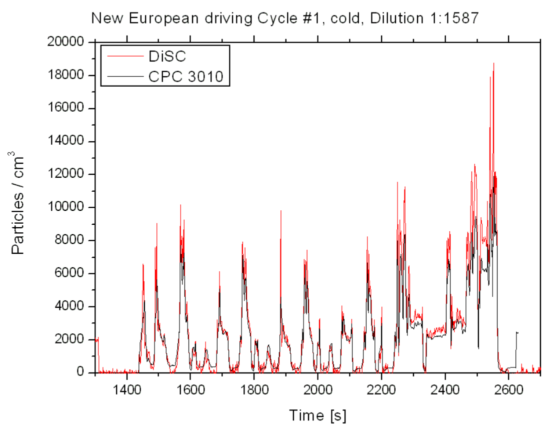

Figure 7: Number concentration measured with a CPC and the DiSC during a New European driving cycle. The same rotating disk diluter (9) is used for both instruments. The result is not corrected for dilution, which is of no interest when comparing the reading of the two devices.

To show the dynamic behavior, the response to a load step in the emissions of a CNG bus is plotted in figure 8. Number concentrations determined by the DiSC, a CPC and an SMPS system as well as the average diameter are shown.

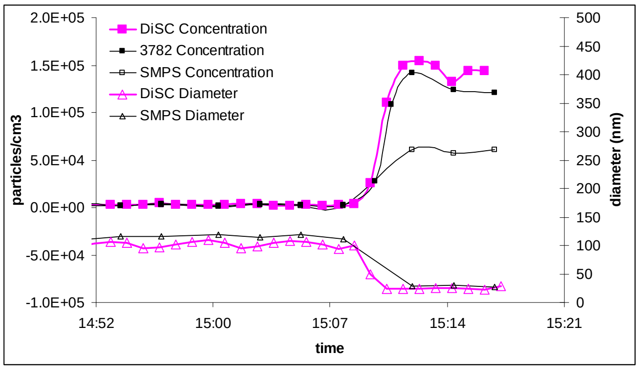

Figure 8: Response of CPC, DiSC and SMPS to a load step of a CNG operated bus. The number concentration and dynamic response obtained from CPC and DiSC are close together. Initial and final average diameter of DiSC and SMPS also match.

To demonstrate the effect of the evaporation tube at the inlet, figure 9 shows results of an ambient air measurement. The measurement location is beside a Swiss highway with heavy traffic which leads to a strong nucleation mode most of the time, except if the wind comes from the direction opposite of the highway. Two DiSCs are operated in parallel, one with and one without evaporation tube. The difference in the number concentration corresponds to the fraction of volatile particles being removed by the evaporation tube.

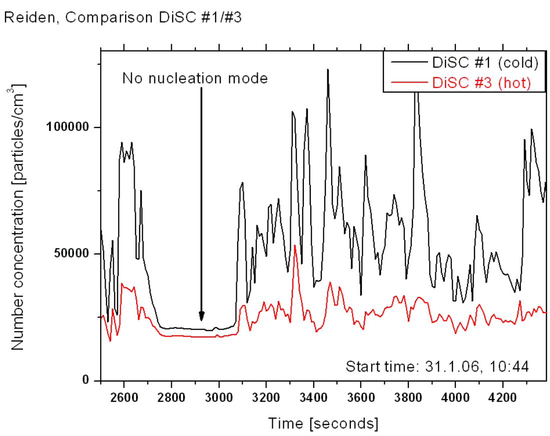

Figure 9: Particle number concentration measured at a highway with heavy traffic with and without evaporation tube. The evaporation tube removes the volatile fraction of the particles. Only when no nucleation mode occurs the number concentration with and without evaporation tube are close together.

Figures 10 and 11 show results for number concentration and average diameter of an ambient air measurement, done outside our lab for a period from Saturday till Monday. DiSC results are compared with number concentration and average diameter calculated from size distributions, obtained with a Scanning mobility Particle Sizer (SMPS, 10) measurements. A TSI 3080 differential mobility analyzer and a TSI 3025 CPC were used. Even if the number concentration obtained by the DiSC is higher the results obtained by the two instruments fit very well.

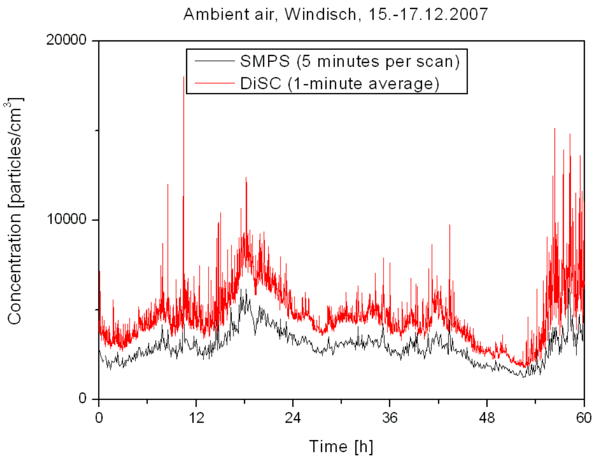

Figure 10: Comparison of number concentration between DiSC and SMPS during an ambient air measurement in Windisch, Switzerland, Starting at Saturday 15.12. 2007, 0:00 and lasting till next Monday,12:00.

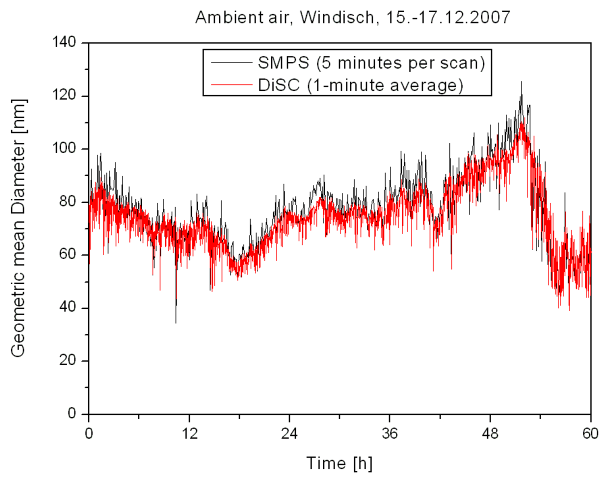

Figure 11: Same measurement as in figure 10, but now the size is compared.

Continuous operation of the DiSC during three weeks beside a highway with heavy traffic did not result in any measurable changes in the performance of the instrument, which might require a new calibration. Also, no visible deposits could be observed. This indicates that the maintenance interval (for calibration, cleaning etc.) is greater that 500 hours of operation at relatively high concentrations.

CONCLUSION

The DiSC is an easily portable and robust device to monitor particle number concentration and average size of nanometer sized particles. The covered size range of 10 - 200 nm is the range of interest for combustion engine emissions as well as for typical urban ambient aerosols. Together with a compact dilution unit, it can be used for a PMP-like measurement in the field The response time of about one second allows transient measurements, for example during transient test cycles or free acceleration.

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REFERENCES

1. Chase, R.E., G.J. Duszkiewicz, J.F.O. Richert, D. Lewis, M.M. Maricq, and N. Xu “PM Measurement Artifact: Organic vapor deposition on different filter media.” *SAE* 2004-01-0967.
2. Official Journal of the European Union "REGULATION (EC) No 715/2007 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 20 June 2007 on type approval of motor vehicles with respect to emissions from light passenger and commercial vehicles (Euro 5 and Euro 6) and on access to vehicle repair and maintenance information" L 171/1, 2007
3. Kasper M. “The number concentration of non-volatile particles, design study for an instrument according to the PMP recommendations.” *SAE* 2004-01-0960.
4. Andersson, J., B. Giechaskiel, R. Muñoz-Bueno, E. Sandbach, P. Dilara "Particle Measurement Programme (PMP) Light-duty Inter-laboratory Correlation Exercise (ILCE_LD) Final Report" www.dieselnet.com, 2007.
5. Baltensperger, U., Weingartner, E., Burtscher, H., and Keskinen, J. "Dynamic mass and surface area measurements.” In K. Willeke, P. A. Baron (Eds.), *Aerosol measurement: Principles, techniques and applications* (pp. 387–418). New York: John Wiley & Sons, 2001.
6. Fierz. M , L. Scherrer, and H. Burtscher "Real time measurement of aerosol size distributions with an electrical diffusion battery.” *J. Aerosol Sci*. 33, 1049-1060, 2002.
7. Keller, A, M. Fierz, K. Siegmann, H.C. Siegmann "Surface science with nanosized particles in a carrier gas" J. Vac. Sci. Technol. A 19(1), Jan/Feb 2001
8. Harris, S., & Maricq, M "The role of fragmentation in defining the signature size distribution of diesel soot. *Journal of Aerosol Science*, *33*, 935–942, 2002.

9. Hüglin, Ch., L. Scherrer and H. Burtscher „An accurate, continuously adjustable dilution system (1:10 to $1:10^4$) for submicron aerosols." *J. Aerosol Sci*. 28, 1049-1055,1997.
10. Wang, S.C. and R.C. Flagan (1990). Scanning electrical mobility spectrometer. *Aerosol Sci. Technol*. 13, 230-240.

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