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CAST Charge Distribution And The Influence Of Neutralizer Performance

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In the framework of the ECE R83/R49 regulations for vehicle homologation, the measurement of particle number concentration in diesel exhaust becomes indispensable. For application with combustion generated nanoparticles, particle counters need to be calibrated with a Combustion Aerosol Standard (CAST). Calibration of condensation particle counters (CPC) with size selected airborne nanoparticles depends on a reliable size selection of the particles. Size selection is mostly done with differential mobility analyzers (DMA) classifying particles according to their electrical mobility *Z*. The presence of particles carrying multiple charges impairs size selection, as larger particles with multiple charges can end up with the same electrical mobility as single charged particles of the desired size. Furthermore, multiply charged particles falsify measurements of particle number concentration obtained with an electrometer.

In the present study, we have investigated the charge distribution of a CAST generated according to Jing (1999). A narrow charge distribution would provide the possibility of obtaining a larger ratio between singly and multiply charged particles as compared to the charge distribution generated by a bipolar charger.

The charge distribution of the CAST aerosol has been measured with a tandem DMA setup. A first DMA selects particles with a specific electrical mobility Z. An SMPS scan of the transmitted particles separates these particles into their different charge levels, so that the number concentration $N(Z,p)$ of particles transmitted by the first DMA can be derived for each charge level p. Comparing $N_0(Z,p)$ for the CAST led into the first DMA without neutralization with $N_n(Z,p)$ for the CAST being neutralized prior to entering the first DMA yields the probability φ_p for a pristine CAST particle to carry p charges, according to

$$\varphi_p\left(d_p(Z)\right)=\psi_p\left(d_p(Z)\right)\cdot\frac{N_0(Z,p)}{N_n(Z,p)}$$

Here, ψ_p is the probability for a neutralized CAST particle to carry p charges, and $d_p(Z)$ is the mobility diameter of a particle with electrical mobility Z carrying p charges. The measured CAST charge distribution is displayed in Fig. 1 for 74 nm and 120 nm mobility diameter, and the equilibrium charge distribution (Wiedensohler, 1988) for the corresponding particle size is shown for comparison.

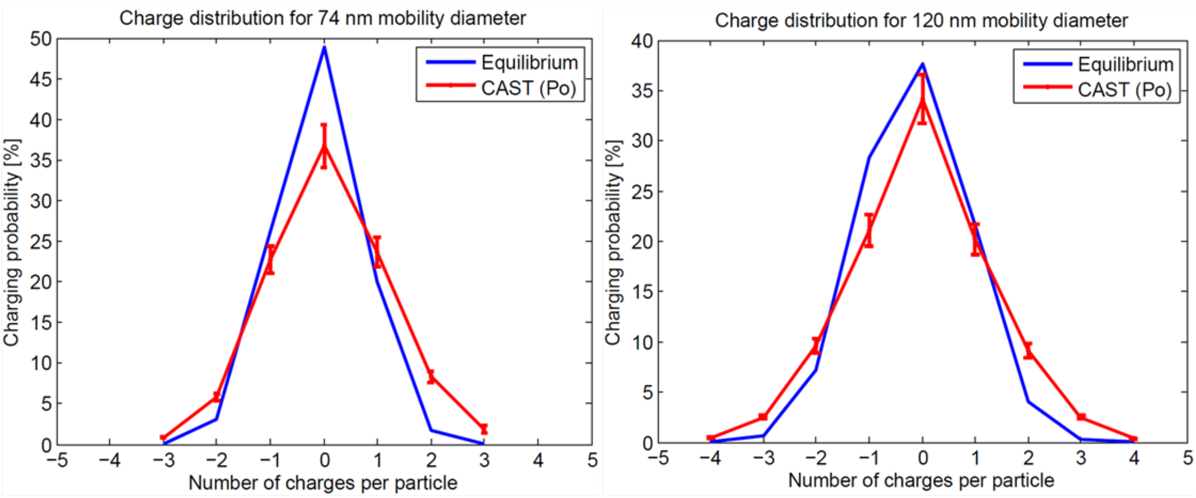

Fig.1: Measured CAST charge distribution as compared to the equilibrium charge distribution

The CAST charge distribution appears to be more symmetric than the equilibrium distribution, and a larger fraction of the particles carries multiple charges. However, absolute differences are quite small. We therefore conclude that concerning the ratio between singly and multiply charged particles, there is no advantage of the CAST charge distribution compared to the equilibrium charge distribution.

During the experiments, the neutralizer in use appeared to be a crucial element to the result of the measured charge distribution. The following issues have been identified:

- For neutralization of the unipolar aerosol leaving DMA 1, the (10 years old) ^{85}Kr neutralizer of DMA 2 needs to be combined with an additional neutralizer in order to reach complete neutralization at a sample flow rate of 1.2 lpm.
- The charge distribution of a neutralized aerosol depends on the neutralizer in use. The measured ratio $R = \psi_{p,+} / \psi_{p,-}$ between the probability for positive and negative *p*-fold charge is displayed in Fig. 3 for an ^{243}Am and a ^{210}Po neutralizer and compared to the equilibrium value of *R*.

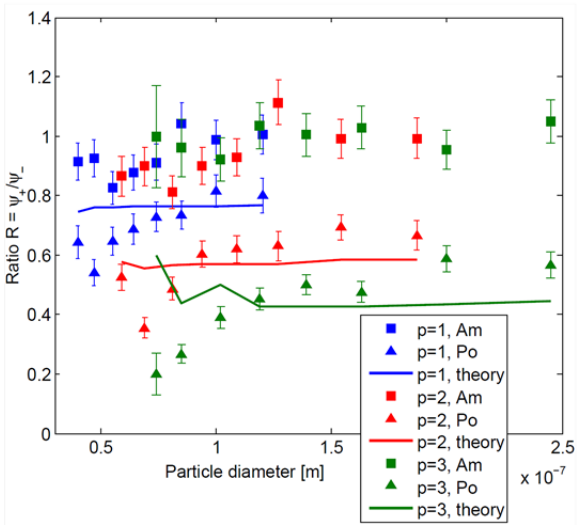

Fig. 2: ^{243}Am Characteristics of a ^{243}Am and a ^{210}Po neutralizer as compared to the equilibrium charge distribution (labeled "theory") for different charge levels p.

Figure 3 shows that for the ^{243}Am neutralizer, *R* is shifted towards more positively charged particles as compared to the equilibrium charge distribution, whereas agreement between measurement and theory is better for the ^{210}Po neutralizer.

We have investigated the potential influence of deviations from the equilibrium charge distribution with a simulated SMPS scan for an ^{243}Am and a ^{210}Po neutralizer under the assumption of equal total number of charged particles. The less pronounced asymmetry in the charge distribution of the ^{243}Am neutralizer as compared to the equilibrium case results in differences between the real size distribution and the distribution measured with an SMPS. For the ^{210}Po neutralizer, this effect is negligible. However, the assumption of equal total number of charges oversimplifies the situation, and the effect of the neutralizer performance may be stronger than reported here.

References

- Jing, L. (1999). Standard combustion aerosol generator (SCAG) for calibration purposes. 3rd ETH Workshop "Nanoparticle Measurement", www.sootgenerator.com
- Wiedensohler, A. (1988). An approximation of the bipolar charge distribution for particles in the submicron size range. J. Aerosol Sci. 19, p. 387 – 389.

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Motivation

In the framework of the ECE R83/R49 regulations for vehicle homologation, the measurement of particle number concentration in diesel exhaust becomes indispensable. For application with combustion generated nanoparticles, particle counters need to be calibrated with a Combustion Aerosol Standard (CAST).

Calibration of condensation particle counters (CPC) with size selected airborne nanoparticles depends on a reliable size selection of the particles. Size selection is mostly done with differential mobility analyzers (DMA) classifying particles according to their electrical mobility *Z*. The presence of particles carrying multiple charges impairs size selection, as larger particles with multiple charges can end up with the same electrical mobility as single charged particles of the desired size.

In the present study, we have investigated the charge distribution of a CAST generated according to Jing (1999). A narrow charge distribution would provide the possibility of obtaining a larger ratio between singly and multiply charged particles as compared to the charge distribution generated by a bipolar charger.

Tandem DMA experimental setup

A tandem DMA setup with two identical DMA's (TSI 3080) has been used (see Fig. 1). DMA 1 selects a specific electrical mobility Z, and an SMPS-scan obtained with DMA 2 provides the number concentration of particles transmitted by DMA 1 for each charge level.

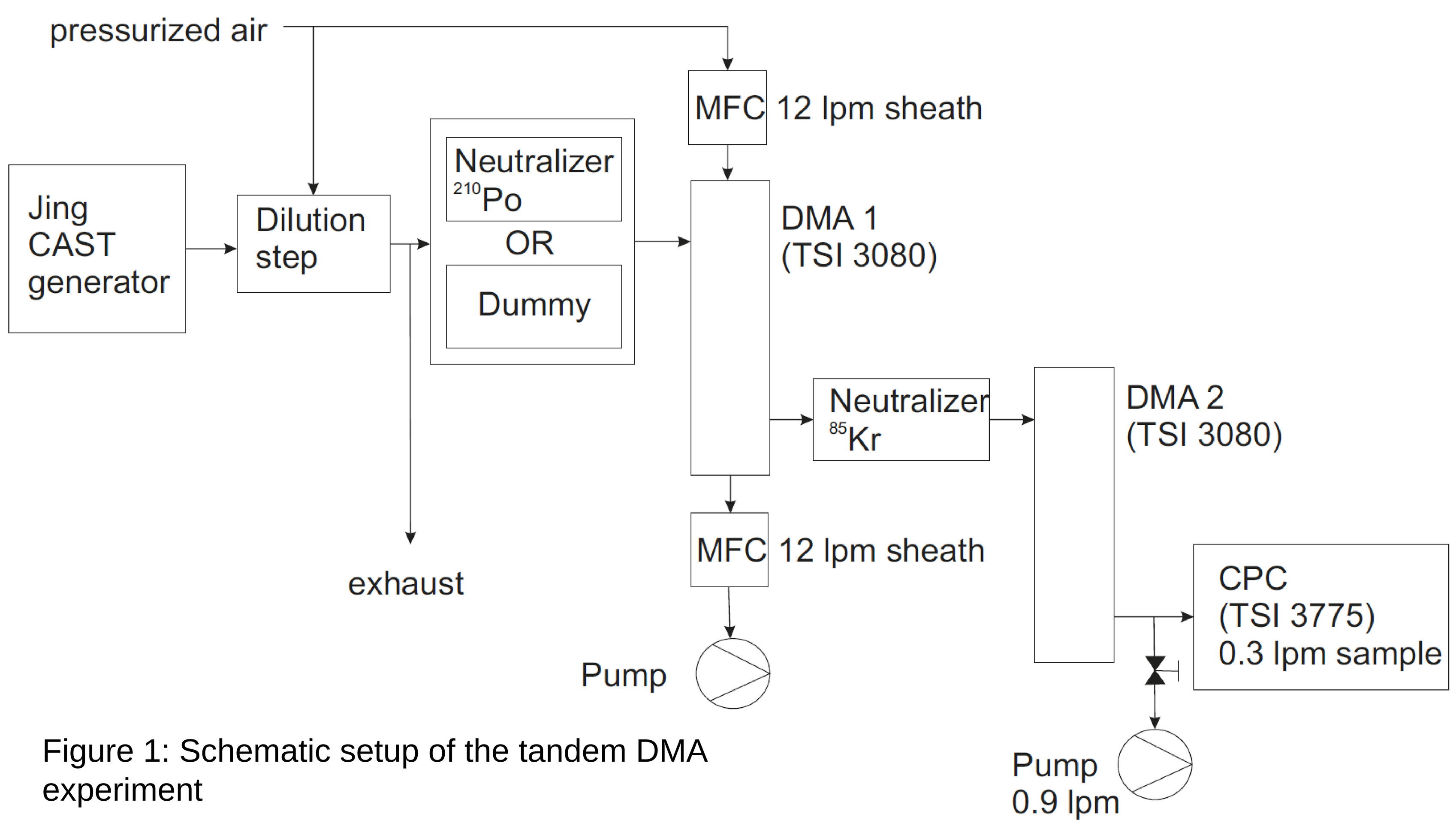

Figure 1: Schematic setup of the tandem DMA experiment

For a fixed *Z* selected by DMA 1, the number concentration of transmitted particles with charge *p* has been determined with a ^{210}Po neutralizer ($N_n(Z,p)$) and with a dummy ($N_0(Z,p)$) in front of DMA 1. The probability $\varphi_p(Z)$ for a particle to carry p charges after passing through the *dummy* can then be calculated according to

$$\varphi_p(Z) = \psi_p(Z) \cdot \frac{N_0(Z,p)}{N_n(Z,p)}$$

where $\psi_p(Z)$ is the probability for a CAST particle to carry *p* charges after passing through the *neutralizer*. Given the relatively high aerosol flow rate (1.2 lpm) and short tubings used, we assume $\varphi_p(Z)$ to be representative for the charge distribution of the particles leaving the CAST generator.

CAST charge distribution vs. equilibrium charge distribution

Figure 2 displays the resulting charge distribution φ_p of the CAST for selected mobility diameters of 74 nm and 120 nm as compared to the equilibrium charge distribution according to Wiedensohler (1988). The CAST charge distribution appears to be more symmetric than the equilibrium distribution, with a tendency to higher probability for multiple charges. However, absolute differences are quite small.

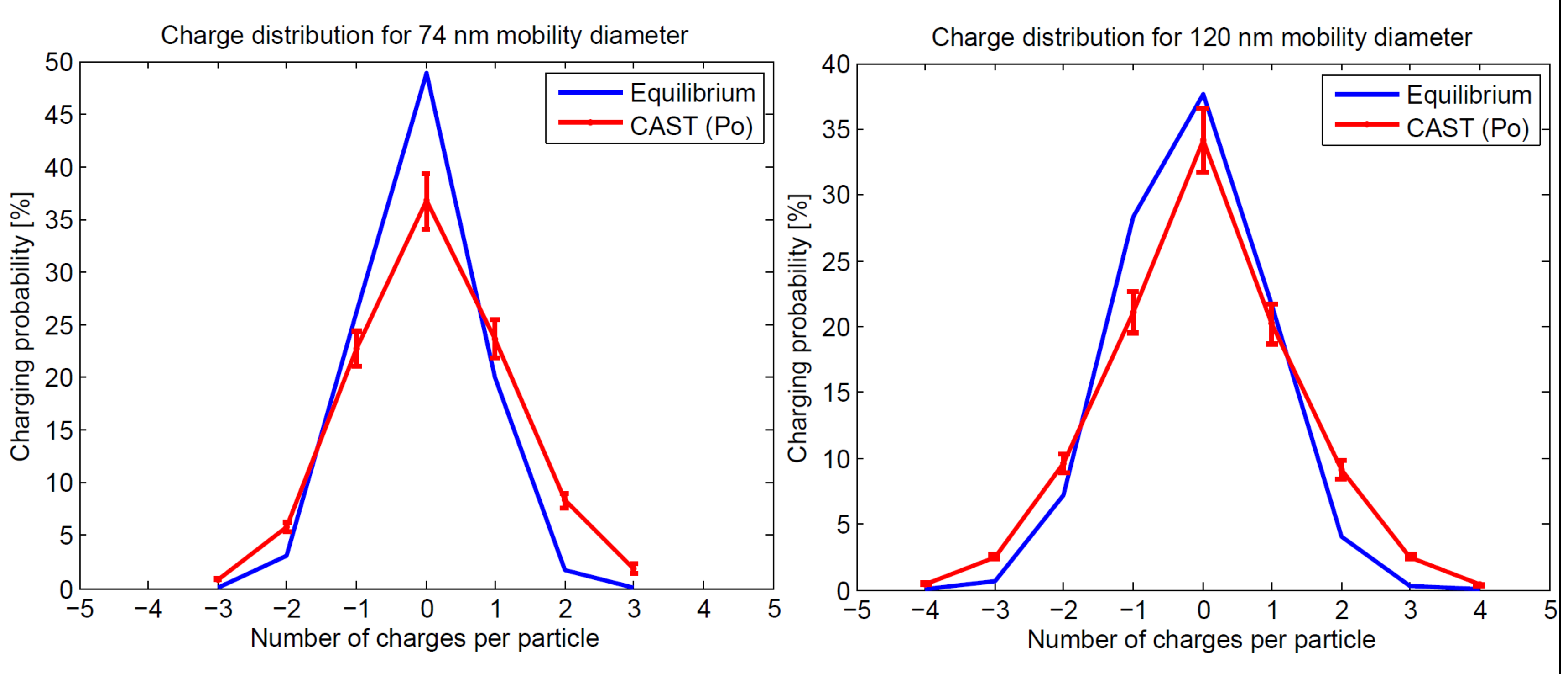

Figure 2: Measured CAST charge distribution as compared to the equilibrium charge distribution for 74 nm and 120 nm mobility diameter.

Neutralizers vs. equilibrium charge distribution

During the experiments, the neutralizers have turned out to be a crucial element to the results of a measurement. The following issues have been identified:

- For neutralization of the unipolar aerosol leaving DMA 1, the (10 years old) ^{85}Kr neutralizer of DMA 2 needs to be combined with an additional neutralizer in order to reach complete neutralization at a sample flow rate of 1.2 lpm.
- The charge distribution of a neutralized aerosol depends on the neutralizer in use. The measured ratio $R = \psi_{p,+} / \psi_{p,-}$ between the probability for positive and negative p-fold charge is displayed in Fig. 3 for an ^{243}Am and a ^{210}Po neutralizer and compared to the equilibrium value of *R*.

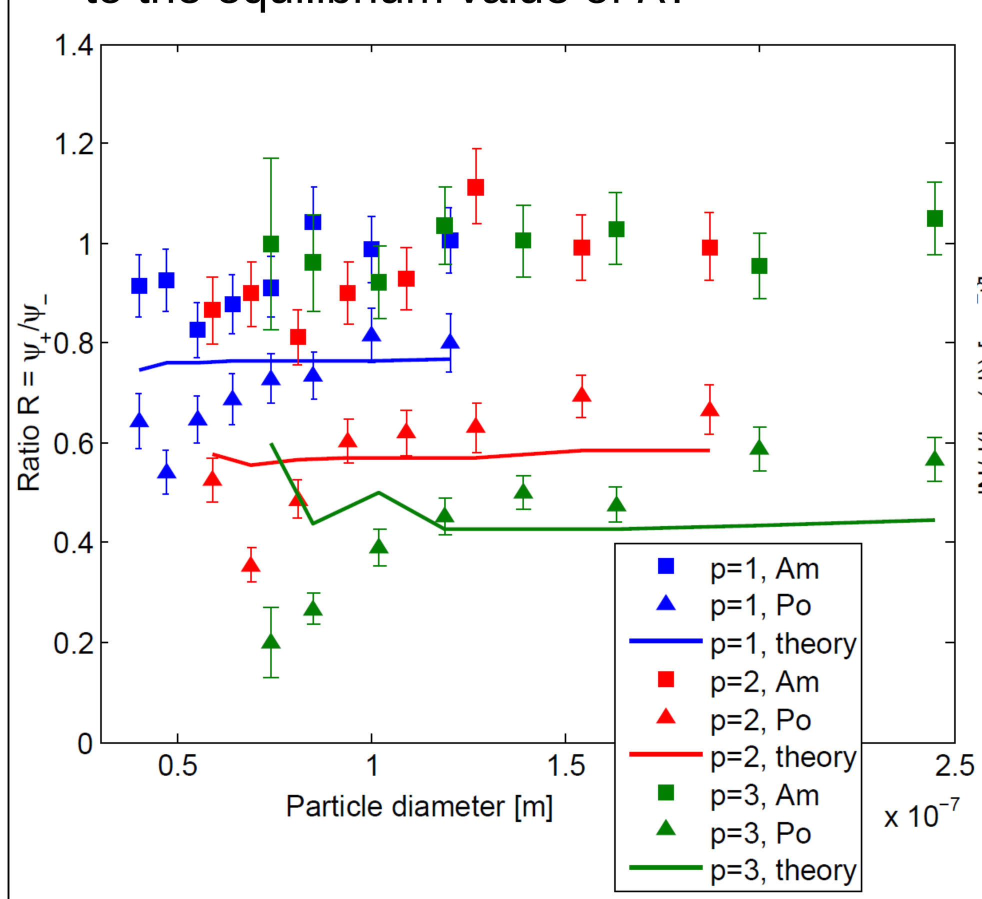

Figure 3: Characteristics of a ^{243}Am and a ^{210}Po neutralizer as compared to the equilibrium charge distribution (labelled «theory») for different charge levels *p*.

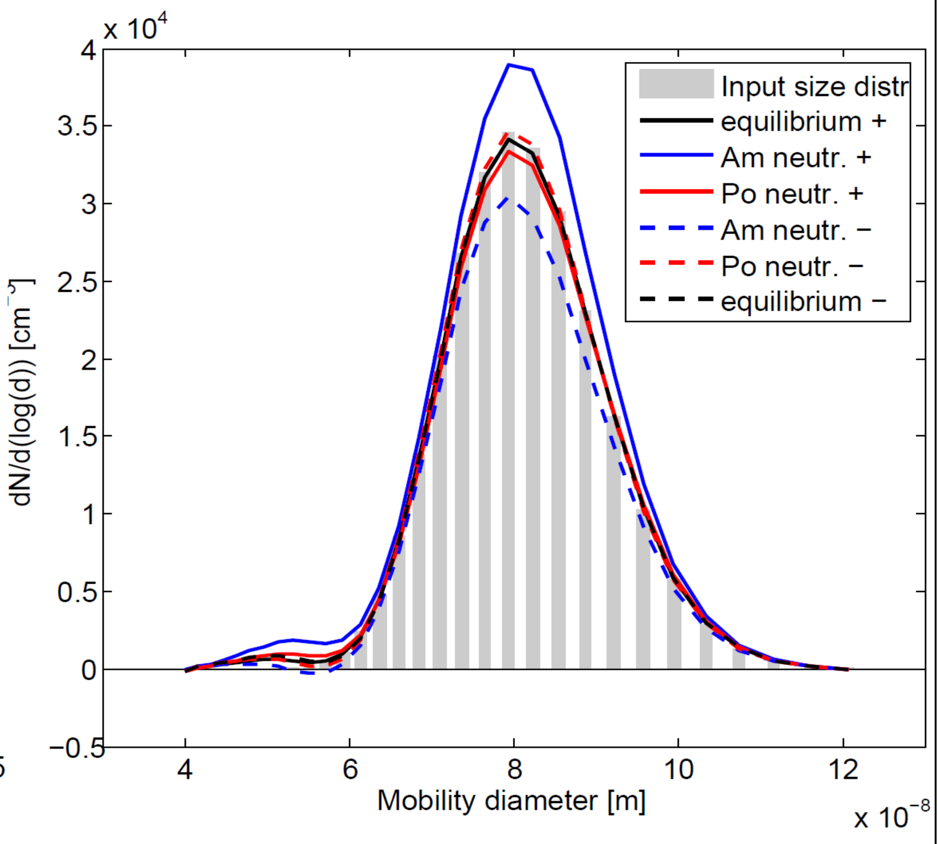

Figure 4: Simulated SMPS scan of a lognormal input size distribution with different neutralizers in front of the SMPS DMA, for positively (+) and negatively (-) particles transmitted by the DMA.

Figure 3 shows that for the ^{243}Am neutralizer, *R* is shifted towards more positively charged particles as compared to the equilibrium charge distribution, whereas agreement between measurement and theory is better for the ^{210}Po neutralizer.

Figure 4 shows the sensitivity of a simulated SMPS scan to the performance of the SMPS neutralizer. The resulting SMPS scan has been calculated for a ^{210}Po neutralizer, an ^{243}Am neutralizer and an equilibrium charge distribution of the aerosol. The total number of charges has been assumed to be equal as in the equilibrium case.

The ^{243}Am neutralizer's charge distribution is more symmetric than the equilibrium charge distribution. This leads to a difference between the real size distribution and the one measured by the SMPS. This effect might be stronger if the total number of charged particles also differs from the equilibrium case.

Conclusions

- The charge distribution of a combustion aerosol produced by a Jing CAST generator is more symmetric and broader than the equilibrium charge distribution. Concerning the ratio between singly and multiply charged particles, the CAST charge distribution has no advantage compared to the equilibrium charge distribution.
- The ^{85}Kr neutralizer of a TSI 3080 SMPS does not fully neutralize an unipolar aerosol from a DMA at 1.2 lpm flow rate.
- Neutralizers do not really generate an equilibrium charge distribution. In the case of the used ^{243}Am neutralizer, this can affect the result of an SMPS scan.

References

- Jing, L. (1999). Standard combustion aerosol generator (SCAG) for calibration purposes. 3rd ETH Workshop "Nanoparticle Measurement", www.sootgenerator.com
- Wiedensohler, A. (1988). An approximation of the bipolar charge distribution for particles in the submicron size range. J. Aerosol Sci. 19, p. 387 – 389.

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