An Approach to a Harmonized Method for Monitoring the Particle Number Size Distribution of Fine and Ultrafine Particles in Ambient Air

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Introduction and Background

The assessment of ambient air quality with respect to the pollution with particulate matter is based on limit values for the mass concentrations PM_{10} and $PM_{2.5}$ [1]. Being aware of the significance of fine and ultrafine particles (D < 1 µm) for human health and for their climatic impact it appears necessary to supplement these mass concentrations with a measurement of the particle number concentration and the particle number size distribution of the fine and ultrafine particles. In 2008, CEN/TC 264 "Air Quality" initiated European standardization projects (Technical Specifications) on the two most relevant measurement techniques in this field: CPC (Condensation Particle Counter) for determining the particle number concentration and MPSS (Mobility Particle Size Spectrometer) for determining the particle number size distribution.

Methodology

The objective of the two Technical Specifications is to define performance characteristics and the associated minimum requirements that enable these instruments (CPC, MPSS), together with a suitable sampling system, to be used in air quality monitoring networks and provide data of adequate accuracy. The first Technical Specification (CEN/TS 16976 [2]) was published in 2016 in the CEN member states. Manufacturers responded quickly and market CPC instruments which meet the requirements. The second Technical Specification (MPSS) is still under development and will presumably be published in 2018/2019.

MPSS Design and Performance Characteristics

Design characteristics	
Pre-separator (optional)	Impactor or cyclone, D_{50} = 1 µm
Bipolar diffusion charger	Bipolar charge distribution according to ISO 15900 [3]
Classifier (DMA)	Cylindrical; p, T, RH measurement; size resolution $\Delta Z/Z \leq 0{,}25$
Particle detector	CPC according to CEN/TS 16976
Size-integrated number concentration range	100 cm^{-3} to 100.000 cm^{-3}
Size resolution of the data deconvolution	≥ 16 geometrically equal distributed size channels per size decade
Time to measure a size distribution	2 min to 10 min
Data availability	ACTRIS level 0, level 1 and level 2
Performance characteristics	
Particle size range	lower limit: 10 nm, upper limit: 800 nm
Calibration accuracy	≤ 3 %
Accuracy of integrated number concentration measurement	$0{,}9 \leq$ slope $\leq 1{,}1$ ($R^2 \geq 0{,}9$)
Mean false background concentration	< 0,01 cm^{-3} for a 10 minute measurement of HEPA filtered air
Size distribution accuracy	Deviation ≤ 10 % for size channels between 20 nm and 300 nm; ≤ 25 % for 10 to 20 nm and 300 nm to 800 nm

MPSS Model Set-up

The schematic of a recommended mobility particle size spectrometer [4] is shown in Fig. 1. The set-up includes dryers for aerosol flow and sheath air, heat exchanger, particle filters, sensors for aerosol and sheath air flow rate, relative humidity and temperature of aerosol flow and sheath air, and absolute pressure.

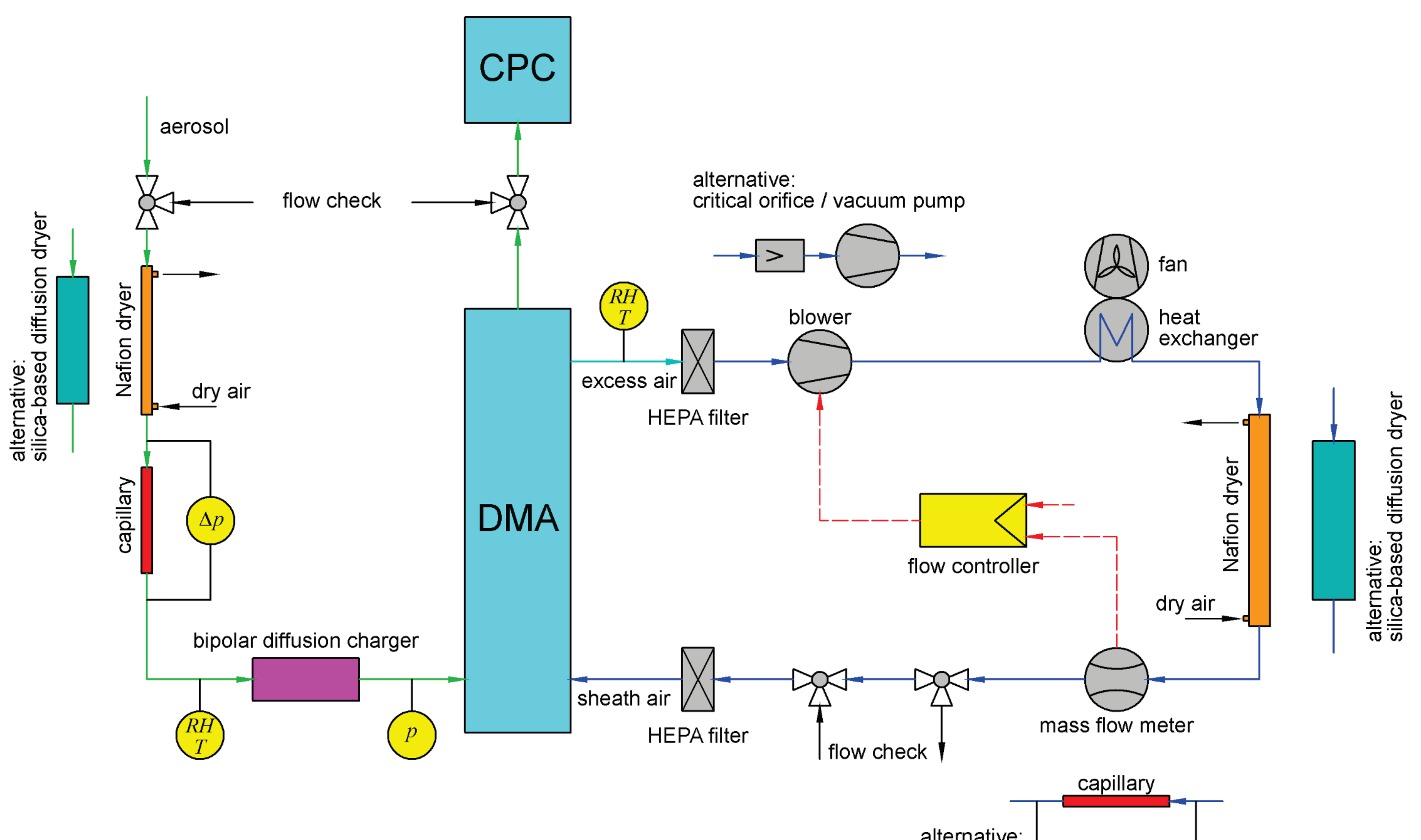

Fig. 1 Schematic representation of a recommended MPSS

Test procedures for the MPSS performance criteria

Particle Sizing Accuracy

The particle sizing accuracy has to be checked experimentally using traceable monodisperse latex aerosols (particle size between 100 nm and 300 nm). If the size given by the measurement is off by 3 % to 10 % from the certified size of the latex particles, the sheath air flow rate of the DMA may be adjusted to achieve correct sizing. Figure 2 shows the result of such a measurement. The large peaks represent the latex particles while the peaks at a smaller particle size are caused by residue particles.

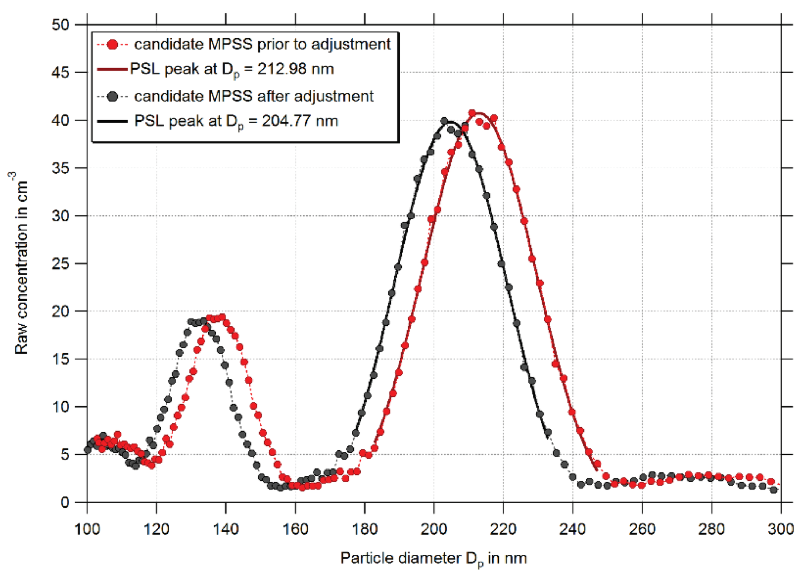

Fig. 2: Particle size distribution (raw concentration) for latex particles of 203 nm certified diameter measured by a candidate MPSS before and after adjustment of the DMA sheath air flow rate

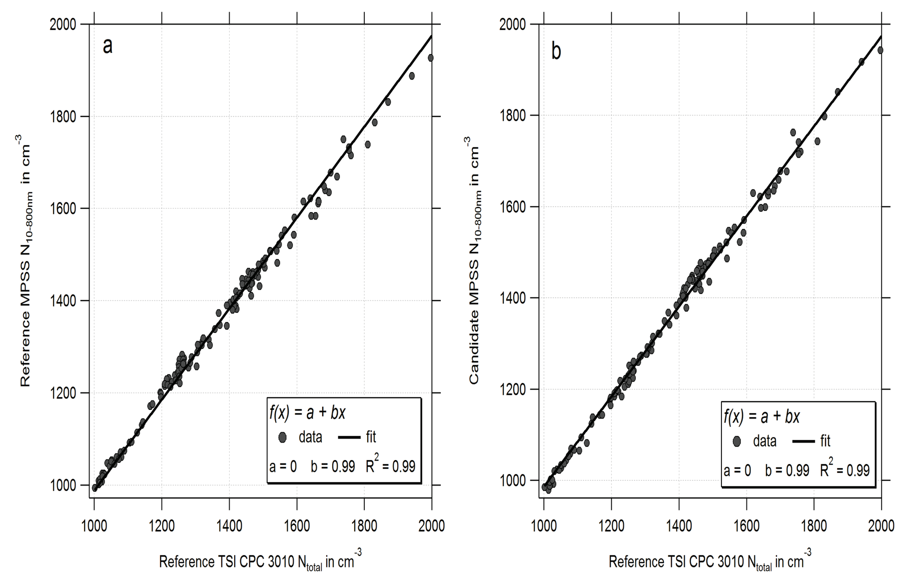

Fig. 3: Comparison of the integrated number concentration of an MPSS with a reference CPC
a) Reference MPSS
b) Candidate MPSS

Accuracy of Integrated Number Concentration Measurement

The accuracy of the total number concentration measurement (number concentration integrated over the MPSS size range) has to be determined experimentally by comparison with the data of a reference CPC. Ambient atmospheric aerosol is used for this comparison. To obtain a reliable counting statistic, the comparison has to cover a period of at least 8 h. The CPC concentrations are averaged over the scan period of the MPSS. The integrated number concentration of the MPSS is plotted against the corresponding CPC concentration. The slope determined by linear regression, forced through the origin, has to meet the requirement given in the Table. Figure 3 shows two examples of these plots.

Size Distribution Accuracy

The size distribution is determined experimentally by comparison with a reference MPSS. Ambient atmospheric aerosol has to be used for this comparison as well. It is recommended to have an additional reference CPC to compare the integrated number concentration of both MPSS with the reading of this CPC. As the number concentration of large particles is rather low, the measured size distributions have to be averaged over a period of at least 8 h to achieve sufficient precision. The comparison has to be done on the basis of these averaged size distributions. The deviation of the size distributions has to be smaller than 10 % in the size range between 20 nm and 500 nm. Figures 4 and 5 give examples for such a comparison. The shaded band represents the allowed tolerance of ±10 %.

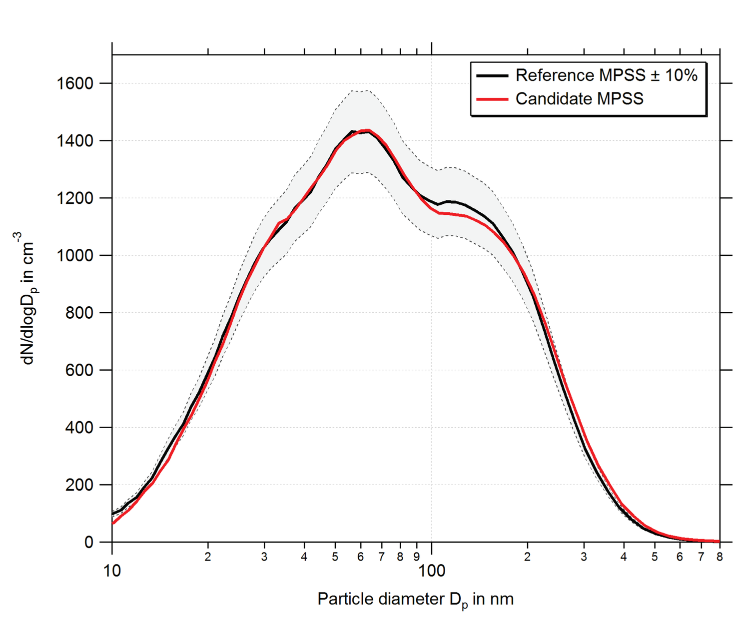

Fig. 4 Comparison of the measured size distributions of a candidate MPSS and of a reference MPSS

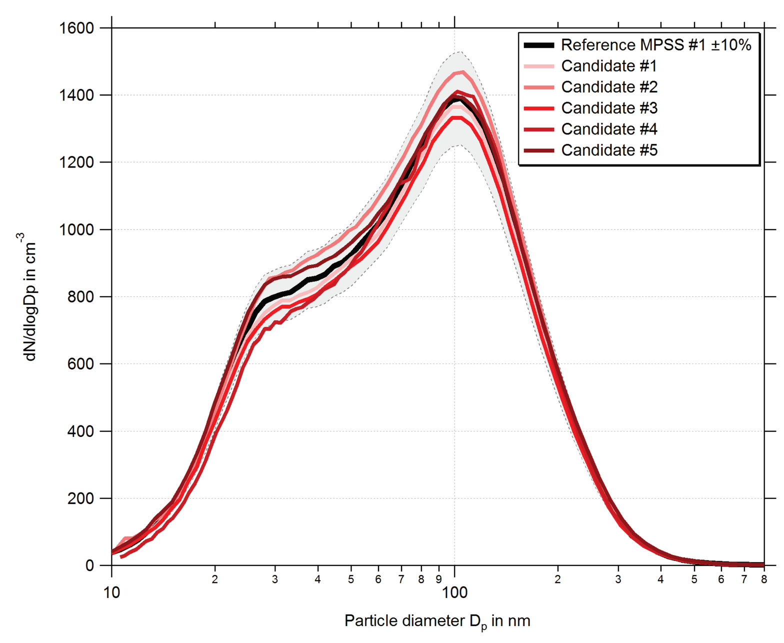

Fig. 5 Comparison of the measured size distributions of five candidate MPSS and of a reference MPSS

Conclusions

The discussion, if it is necessary to implement limit values for the particle number concentration (and even including the particle size distribution) in ambient air, is in progress. The main counter-argument is that the results of long-term epidemiologic studies which demonstrate the evidence of human health effects with sufficient significance, are still missing. If such results are once available the concept and infrastructure of air quality monitoring can then be based on existing European Standards.

References

[1] Directive 2008/50/EC on ambient air quality and cleaner air for Europe
[2] CEN/TS 16976 CPC Ambient air – Determination of the particle number concentration of atmospheric aerosol (2016)
[3] ISO 15900 Determination of particle size distribution – Differential electrical mobility analysis for aerosol particles (2009)
[4] Wiedensohler, A., et al.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmos. Meas. Tech., 5 (2012) 657–685