Journal of Aerosol Science 68 (2014) 14–24

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

An ultrafine particle monitor for size-resolved number concentration measurements in atmospheric aerosols

L. Hillemann [a,*], A. Zschoppe [b], R. Caldow [c], G.J. Sem [c,1], A. Wiedensohler [d]

[a] Institute of Process Engineering and Environmental Technology, TU Dresden, 01062 Dresden, Germany
[b] Topas GmbH, Oskar-Röder-Str. 12, 01237 Dresden, Germany
[c] TSI Incorporated, 500 Cardigan Road, Shoreview, MN 55126, USA
[d] Leibniz Institute for Tropospheric Research, Permoser Str. 15, 04318 Leipzig, Germany

ARTICLE INFO

Article history:
Received 26 April 2013
Received in revised form
24 October 2013
Accepted 25 October 2013
Available online 4 November 2013

Keywords:
Particle number concentration monitor
Atmospheric aerosol
Corona-jet charger
Kernel matrix

ABSTRACT

An ultrafine particle monitor (UFPM) was designed and constructed to obtain size-resolved particle number concentrations of atmospheric aerosols in air quality monitoring networks. The UFPM employs a corona-jet charger for particle charging, a DMA for electrical mobility classification, and an aerosol electrometer for particle quantification based on electrical charge. It does not require a radioactive source or an alcohol-based particle detector. A sensor model of the UFPM was derived, which relates the measured electrical current to the particle number concentration. A data inversion algorithm based on the Tikhonov-regularization was implemented. It yields the number concentration in six size classes between 20 and 720 nm from a 10-min-measurement with an upper limit of $3\times10^{6}\,\mathrm{cm}^{-3}$. A method was developed to calculate the kernel matrix from the data of comparison measurements with the UFPM and a reference mobility particle size spectrometer such as an SMPS. In experimental comparisons, the number concentrations of laboratory and atmospheric aerosols measured by the UFPM were within a range of $\pm20\%$ of the reference concentration determined by an SMPS.

1. Introduction

Depending on particle size distribution and chemical composition, atmospheric aerosols have various impacts on human health. Epidemiological studies indicate a correlation between fine and ultrafine (<100 nm) particle exposure and respiratory and cardiovascular mortality (Katsouyanni et al., 2001; Samet et al., 2000). This correlation is highly significant, if the particle exposure is traffic-generated (Rosenlund et al., 2006) and it is stronger for PM2.5 than for PM10 (Dockery et al., 1993; Schulz et al., 2005). These studies suggest that especially ultrafine particles are responsible for the negative impact of aerosols on human health (Brüske-Hohlfeld & Peters, 2010). Although the mechanisms of these effects are not entirely understood, the size effect is induced by the high deposition of ultrafine particles in the deep lung (Hinds, 1999).

Due to the adverse health effects of particle exposure, the particle number concentration in the atmosphere is quantified in air quality monitoring networks. For this purpose, more than 400 monitoring stations exist in Germany (Zellner, 2011).

* Corresponding author. Tel.: +49 35146332914; fax: +49 35146337058.
E-mail address: lars.hillemann@tu-dresden.de (L. Hillemann).
[1] Retired.

0021-8502/$ - see front matter
http://dx.doi.org/10.1016/j.jaerosci.2013.10.007

Currently, particle concentrations are legally defined as PM10 mass concentrations. Unfortunately, ultrafine particles contribute insignificantly to the aerosol mass concentration and cannot be quantified separately by conventional mass measurement techniques. They can be characterized more accurately by their number concentration (Peters et al., 1997). This exposure metric is already used for atmospheric air quality monitoring in research infrastructure networks such as GUAN (German Ultrafine Aerosol Network) and ACTRIS (Aerosol, Cloud and Trace Gases Research Infrastructure Network) (Asmi et al., 2011; Wiedensohler et al., 2012). They provide highly quality-assured measurement data for scientific purposes. Due to the high maintenance costs, for routine exposure monitoring a simpler measurement system is required (Löschau et al., 2010).

The implementation in monitoring stations defines two operational constraints for particle number concentration monitors. The instruments may not use radioactive sources, due to the stringent safety regulations for the transport of these materials. Furthermore, they should not contain an alcohol-based CPC, because its emission may interfere with hydrocarbon concentration measurement in the monitoring station.

For the measurement of ultrafine particle number concentrations, several mobility particle size spectrometers have been designed and used. These instruments are based on the classification of particles in a DMA or a low-pass mobility filter. Upstream the classifier the charge distribution of the particles is controlled by a radionuclide-based bipolar diffusion charger or a corona-discharge-based unipolar diffusion charger. For the particle quantification a CPC or an aerosol electrometer is employed (Flagan, 1998, Intra & Tippayawong, 2008). The following combinations have been commercially produced:

- a corona-discharge-based unipolar diffusion charger, a low-pass mobility filter and an aerosol electrometer (electrical aerosol analyzer: Liu et al., 1974, Whitby & Clark, 1966);
- a radionuclide-based bipolar diffusion charger with a DMA and CPC (DMPS: Fissan et al., 1983; Ten Brink et al., 1983; SMPS: Wang & Flagan, 1990);
- a radionuclide-based bipolar diffusion charger with a DMA and an aerosol electrometer (electromobility spectrometer: Han et al., 2000; Seol et al., 2000; Shimada et al., 2005; Winklmayr et al., 1991).

Additionally, several custom-designed mobility particle size spectrometers have been designed for scientific purposes as described in Wiedensohler et al. (2012).

The combination of a corona-discharge-based unipolar charger with a DMA and an aerosol electrometer satisfies the constraints discussed above. However, this has been realized up to now only in multichannel electrical aerosol spectrometers (fast mobility particle sizer spectrometer, engine exhaust particle sizer spectrometer: Mirme & Peil, 1983, Mirme & Tamm, 1991, Johnson et al., 2003, differential mobility spectrometer: Biskos et al., 2003, 2005, electrical aerosol spectrometer: Tammet et al., 2002), which are not suited for monitoring stations for economic reasons.

The objective of the present study is to examine, if the combination of a corona-discharge-based unipolar charger with a DMA and only a single aerosol electrometer can be used to quantify the aerosol number concentration in monitoring stations. The results were used to design the ultrafine particle monitor (UFPM). The following sections describe design, construction and the data inversion of the UFPM as well as the results of comparison measurements to a reference instrument.

For implementation in a monitoring station, an UFPM has three operational requirements:

- The particle number concentration should be measured from 20 to at least 500 nm with a reasonable number of size classes.
- The particle number concentration should cover the range from 10^3 to 10^6 cm^{-3}.
- The deviation of the particle number concentration compared to the reference instruments should be lower than $\pm 20\%$.

The UFPM was commercialized by TSI Inc. as model 3031.

2. Description of the ultrafine particle monitor

Figure 1 is a schematic diagram of the UFPM. The aerosol sample passes through a corona-jet charger, which charges all particles positively by diffusion charging. Subsequently, the charged particles are classified in a DMA according to their electrical mobility and quantified by a Faraday cup aerosol electrometer. The sheath flow loop of the DMA consists of a blower, a heat exchanger, a flowmeter and HEPA-filters. A flow controller maintains a constant flow through the sheath flow loop. The heat exchanger is used to remove heat added to the flow loop by the blower. The flow through the aerosol electrometer is maintained by the sample pump. The filter after the sample pump prevents the emission of instrument-generated particles.

Figure 2 shows the corona-jet charger, employed in the UFPM (Intra & Tippayawong, 2009, Medved et al., 2000, Whitby & Clark, 1966). The unipolar corona discharge ion generator produces positive ions by corona discharge in a needle–orifice configuration (Whitby, 1961). The corona current is adjusted to 1 μA at a corona voltage of typically 2.4 kV. The ions are