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A concept of an automated function control for ambient aerosol measurements using mobility particle size spectrometers

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Abstract

An automated function control unit was developed to regularly check the ambient particle number concentration derived from a mobility particle size spectrometer as well as its zero-point behaviour. The aim of the new feature is to conduct unattended quality control experiments under field conditions at remote air quality monitoring or research stations. The automated function control also has the advantage of being able to get a faster system stability response than the recommended on-site comparisons with reference instruments. The method is based on a comparison of the total particle number concentration measured by a mobility particle size spectrometer and a condensation particle counter removing the diffusive particles approximately smaller than 25 nm in diameter. In practice, the small particles are removed by a set of diffusion screens, as traditionally used in a diffusion battery. The other feature of the automated function control is to check the zero-point behaviour of the ambient aerosol passing through a high-efficiency particulate air (HEPA) filter.

An exemplary one-year data set is presented for the measurement site Annaberg-Buchholz as part of the Saxon air quality monitoring network. The total particle number concentration derived from the mobility particle size spectrometer overestimates the particle number concentration by only 2 % (grand average offset). Furthermore, tolerance criteria are presented to judge the performance of the mobility particle size spectrometer with respect to the particle number concentration. An upgrade of a mobility particle size spectrometer with an automated function control enhances the quality of long-term particle number size distribution measurements. Quality assured measurements are a precondition for intercomparison studies of different sites. Comparable measurements will improve cohort health and also climate-relevant research studies.

1 Introduction

For more than three decades, measurements of particle number size distributions (PNSD) have been employed in atmospheric science to investigate the evolution of aerosol particles in time and space worldwide (Bates et al., 1998; Heintzenberg, 2009; Huebert et al., 2003; Raes et al., 2000). The PNSD is a basic parameter needed, e.g. to calculate atmospheric particle light scattering effects and also to estimate the number of cloud condensation nuclei (Romakkaniemi et al., 2012). The number concentration of particles originating from secondary formation has been recognized as playing a certain role in a feedback mechanism moderating climate change (Paasonen et al., 2013). The PNSD also serves as the most basic parameter to describe aerosol particles in atmospheric chemistry transport models and has already been implemented in the European research infrastructure networks EUSAAR (European Supersites for Atmospheric Aerosol Research) and ACTRIS (Aerosols, Clouds and Trace Gases Research Infrastructure Network) (Asmi et al., 2011) and in the German Ultrafine Aerosol Network (Birmili et al., 2009).

Additional motivation to incorporate particle number measurements in air quality monitoring networks derives from the wish to better characterize the health-related properties of atmospheric particles (Russell and Brunekreef, 2009; Wichmann et al., 2000). Furthermore, PNSD measurements in air quality monitoring networks are currently used as a complementary part of legal PMx metrics (particulate matter $< 10\,\mu m$ and $< 2.5\,\mu m$) to interpret the success of clean air plans in congested urban areas (Löschau et al., 2013; Löschau et al., 2012).

The Saxon State Office for Environment, Agriculture and Geology (LfULG) is one of the initiators to integrate PNSD measurements into Germany's Saxon air quality monitoring network. Saxony is situated in the south-eastern part of Germany, sharing borders with Poland and the Czech Republic. Currently, three stations of the Saxon air quality monitoring network are equipped with mobility particle size spectrometers designed by the Leibniz Institute for Tropospheric Research (TROPOS), Leipzig, Germany: the roadside station Dresden-Nord (EU-code DESN061), the urban background station Dresden-Winckelmannstrße (EU-code DESN092), and the urban background station Annaberg-Buchholz (EU-code DESN001). Detailed information about the stations can be found by using the EU-codes on the web page of the Federal Environment Agency of Germany (UBA, 2013).

A regular quality assurance program, which means a connection to a reference, is vital for the comparability of multi-city PNSD measurements. For the quality control of a PNSD measurement, two parameters must be checked: the particle number concentration (PNC) and the particle size. The correct sizing of the mobility particle size spectrometer can be checked by using certified size standards (e.g. NIST[1] certified polystyrene size standards). A particle number concentration standard is nonexistent at the current state-of-the-art. Thus, the direct verification of the correct count rate of a mobility particle size spectrometer is not possible. In this work, we present an operational quality control using a reference method.

The LfULG in cooperation with the Leibniz Institute for Tropospheric Research (TROPOS) developed an automated function control and tested it for suitability in an air quality monitoring network. The method is based on performing a comparison between the mobility particle size spectrometer and a condensation particle counter (CPC) as a reference method for the total PNC. In recent years, on-site comparisons with a reference instrument were done approximately every three to four times a year. One reason to establish the automated function control was to reduce the effort of frequent on-site comparisons and thus the operating expenses. Furthermore, the automated function control shortens the time span of quality checks, which results in a faster intervention in case of problems.

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2 Instrumental

Figure 1 illustrates a schematic sketch of a mobility particle size spectrometer that is equipped with the automated function control. The basic closed-loop mobility particle size spectrometer shown on the right of the sketch fulfils the ACTRIS recommendations (Wiedensohler et al., 2012). All parts of the automated function control are confined by a red dashed line. The purpose of the uppermost motor-driven ball valves is to switch the aerosol flow between (a) ambient aerosol, (b) ambient aerosol passing through a high-efficiency particulate air (HEPA) filter (referred to as zero air), and (c) ambient aerosol passing through a set of 40 diffusion screens. Technical data and design of the diffusion screens are given further down in this paragraph. Downstream of the bypass, the aerosol flow is split into 1 L min^{-1} for the mobility particle size spectrometer and for the transfer CPC, in that order.

The transfer CPC is a commercial condensation particle counter (CPC model 3772, TSI Inc., Shoreview, MN, USA) that serves as a transfer standard, and is regularly cycled between the three measurement stations and the calibration facility (WCCAP – World Calibration Center for Aerosol Physics hosted at the Leibniz Institute for Tropospheric Research). The usual procedure is to operate the transfer CPC at a station for about two weeks, and then proceed to the next measurement site. Two weeks correspond with the standard maintenance cycle in the air quality monitoring network performed by the station personnel.

After each cycle, the total PNCs of the transfer CPC and a stationary master CPC are compared at the WCCAP. Beyond that, the transfer CPC is calibrated against the reference aerosol electrometer (model 3068B, TSI Inc., Shoreview, MN, USA) at the WCCAP once a year. The aerosol electrometer is calibrated at the German metrology institute (PTB) against a well-defined femtoampere source. The calibration of the transfer CPC comprises the determination of the detection efficiency curve of the CPC from 5 up to 40 nm in particle diameter (D_p). Details about the calibration procedure can be taken from the ACTRIS recommendations (Wiedensohler et al., 2012). Figure 2 shows the result of the calibration of the transfer CPC on 24 October 2012. For $D_p < 20$ nm, the detection efficiency curve of the CPC drops steeply, while the current 50 % detection efficiency diameter D_{p50} is 7.2 nm (For comparison, the CPC user manual indicates $D_{p50} = 10$ nm). For $D_p > 30$ nm, the asymptotic value of the detection efficiency of the transfer CPC approaches 100 %. These results might also have been reproduced in a further calibration on 8 August 2013 (not shown here).

Mobility particle size spectrometers have, however, larger uncertainties than CPCs for $D_p < 20$ nm due to deviations in precise high voltage power supply especially for very low DMA voltages. Moreover, DMA characteristics, e.g. material used for the insulator, lead to additional losses, which are not considered in the data processing described in Sect. 3. Therefore, a main objective of the automated function control with diffusion screens is the elimination of such uncertainties, which appear when total PNCs derived from mobility particle size spectrometers and CPCs are compared. The purpose of the diffusion screens is, thereby, the removal of particles at the lower size distribution end and the reduction of the total particle number concentration.

Figure 3 illustrates the diffusion screens (as traditionally used in a diffusion battery) with a diameter of 47 mm and the prototype of the screen holder. The custom-made screen holder is made of two stainless steel pieces. Both pieces adhere together via a sleeve and can be opened by a thumbscrew to take out the screens. The commercial diffusion screens (Drahtweberei Pausa GmbH, Pausa/Vogtland, Germany) are made of stainless steel and have a mesh size of 50 µm and a wire diameter of 35 µm. The screens are enclosed by a mesh and an o-ring with an inner diameter of 40 mm to fix the screens inside the screen holder.

The upper graph of Fig. 4 illustrates an intercomparison of integrated PNC from mobility particle size spectrometer (abbr. $\mathrm{PNC_{mob.}}$) and PNC from the transfer CPC (abbr. $\mathrm{PNC_{tCPC}}$). These data were aggregated from the urban background station Annaberg-Buchholz. For PNC > 5000 $\mathrm{cm^{-3}}$, the scatter plot deviates from the 1 : 1 line, in particular $\mathrm{PNC_{mob.}}$ underestimates $\mathrm{PNC_{tCPC}}$. The underestimation is highest during daytime (cf. lower graph in Fig. 4) when nucleation mode particles originated mainly from traffic and photochemical processes are present. Thus, $PNC_{mob.}$ and PNC_{tCPC} would be much more comparable if nucleation mode particles were removed.

Figure 5 illustrates the two PNSD with and without diffusion screens. It is worth mentioning that both size distributions are measured at the same time. Black crosses in Fig. 5 represent the entire transmission of the diffusion screens, determined as the ratio of both PNSDs. Particles smaller than $D_p = 20$ nm are completely removed by the diffusion screens. Thus, comparisons such as $PNC_{mob.}$ vs. PNC_{tCPC} are less prone to fluctuations in the ambient aerosol in that size range. For particles larger than $D_p = 20$ nm, the transmission of the diffusion screens increases and culminates at about 80 % for the largest particles.

3 Data processing

The following section describes the data processing of the zero air and diffusion screen measurements starting from the raw data. The raw data set comprises the electrical particle mobility distribution covering a mobility diameter range from 10 to 800 nm, as well as diagnostic parameters, i.e. temperature, relative humidity, and flow rates of the aerosol and sheath air. Information about status parameters (0 and 1) for zero air measurements, diffusion screen measurements, and service maintenance are also provided.

The processing of the electrical mobility distribution into the true PNSD contains multiple charge correction (Pfeifer et al., 2013), coincidence correction of the CPC, and correction for diffusional losses in the mobility particle size spectrometer (Wiedensohler et al., 2012). The correction of the counting efficiency of the CPC was not applied for comparisons with the PNC derived from a CPC (here: transfer CPC). A precondition for that is that both CPCs should have the same detection efficiency, which is true in our case. During data processing, the diagnostic parameters were used for automatic flagging and rejection of the respective data set. The range for valid data are 10–30 °C for temperature, 0–40 % for relative humidity, ±2 % of the set point value for the sheath air flow rate, and ±5 % of the set point value for the aerosol flow rate according to the recommendations (Wiedensohler et al., 2012). With regard to the status parameters, the entire data set was classified automatically into data sets for regular measurements without service maintenance, zero air measurements, as well as diffusion screen measurements. The first and the last 5 min of each zero air measurement, typically referred as blanking time, were disregarded. The PNSD measured with diffusion screens were integrated over the entire size range yielding $\mathrm{PNC}_{\mathrm{mob.}}$. Finally, these new data sets were averaged (arithmetic mean) to a lower time resolution of 1 h.

4 Results and discussion

The following section depicts exemplary results of the automated function control for the Annaberg-Buchholz site after necessary software and hardware upgrades of the mobility particle size spectrometer were completed.

Figure 6 shows results of diffusion screen measurements for a full year, from 1 August 2012 to 1 August 2013. The upper graph presents the ratio of $\mathrm{CPC}_{\mathrm{mob.}}$ to $\mathrm{CPC}_{\mathrm{tCPC}}$ for each single diffusion screen measurement (black crosses) and the mean value of the entire period (red solid line). The mean value is 1.02 (offset of 2 %) and there is no significant trend visible.

Nevertheless, variability can be seen from cycle to cycle, which cannot be fully explained at present. An influence of $\mathrm{PNC}_{\mathrm{tCPC}}$, which is illustrated in the lower graph, can be excluded. Obviously, $\mathrm{PNC}_{\mathrm{tCPC}}$ does not correlate with the ratio. Besides the cycle to cycle variability, there is a certain intra-cyclic variability as well, which is caused by the automatic function control setup itself. In particular, the transfer CPC evaluates all size classes simultaneously, whereas the mobility particle size spectrometer measures the size classes successively. Fast variations in PNC or fast variations in the shape of the PNSD cannot be detected by the automated function control. A special case of intra-cyclic variability occurred during March 2013. Here, the focusing inner nozzle of

Title Page
Abstract Introduction
Conclusions References
Tables Figures
I◀ ▶I
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

the CPC as part of the mobility particle size spectrometer became increasingly clogged accompanied by a decrease of the particle counting efficiency.

An aim of this work is to derive tolerance criteria based on the diffusion screen and zero air measurements of the previous year. These tolerance criteria will help to judge the instrument performance in future and can be stringently applied to judge data valid or invalid. The conduction of tolerance criteria for the diffusion screens using error propagation would lead to unrealistic errors and is therefore impracticable. Instead, we employed a statistical approach, which is also used for equivalence checks of reference instruments in air quality monitoring networks. There, the mean value ± three standard deviations (σ) is applied to rate the performance of a measurement device. For our exemplary data set, the mean $\pm 3\sigma$ value is 1.02 ± 0.19 (cf. upper graph in Fig. 6). Thus, the tolerance criteria for the mobility particle size spectrometer at the Annaberg-Buchholz site would be 0.83 and 1.21, respectively. Presumably, these tolerance criteria are site specific.

In practice, these tolerance criteria could be applied as follows: if the mean value of the diffusion screen measurements of one cycle (duration typically 2 weeks) drops below or exceeds the tolerance criteria, further inspections should follow in order to fix the problem. As a consequence, the regular ambient aerosol measurements would be invalidated as long as the subsequent diffusion screen measurement turns into the valid range.

Table 1 presents condensed results from the zero air measurements for seven size ranges (10–20 nm, 20–30 nm, 30–50 nm, 50–70 nm, 70–100 nm, 100–200 nm, and 200–800 nm) including minimum, maximum, and entire mean values. The highest PNC of 2 cm^{-3} was recorded for the smallest size class of 10–20 nm. The establishment of a tolerance criterion for the zero air is based on the lower detection limit for the PNC and the permeation rate of the zero filter. The lower detection limit is dependent on the false count rate of the CPC, which is in turn so low that the resultant lower detection limit is approximately zero. The permeation rate of the used HEPA filter is approximately 5×10^{-3} for 300 nm sized particles. With regard to common particle concentrations measured at ambient conditions, we suggest a tolerance criterion for the PNC of each size range that is 2 cm^{-3}.

5 Summary and conclusion

This paper presents a new concept in the quality control for long-term measurements with mobility particle size spectrometer. Quality assured measurements are a precondition for intercomparisons of different sites. The automated function control fulfils the requirements of measurement networks for monitoring the system performance quasi-continuously. The automated function control comprises diffusion screen and zero air measurements and checks the instrument performance with respect to the particle number concentration. The special feature of the automated function control is its ability to get a faster response on the system stability than recommended on-site comparisons with reference instruments from the calibration facility. A major advantage of the automated function control is the software-driven switching between regular ambient air, zero air, and diffusion screen measurement. Moreover, the maintenance work is quite low. Beside the setting up and dismounting of the transfer CPC, the diffusion screens should be regularly cleaned in an ultrasonic bath and the filter cartridge has to be replaced from time to time. The mobility particle size spectrometer has to be upgraded by one further CPC and some hardware parts and the software must be updated.

The performance of a mobility particle size spectrometer equipped with an automated function control is exemplary shown for the measurement site Annaberg-Buchholz for the one-year period 1 August 2012 to 1 August 2013. For the particle number concentration, an offset of 2 %, but no significant trend, was found from the comparison with the transfer CPC for the one-year period. Statistical derived tolerance criteria were calculated to 0.83 and 1.21 for the same period. For the zero air measurement, a tolerance criterion for the particle number concentration of 2 cm^{-3} is suggested.

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Table 1. Results of the zero air measurements from 1 August 2012 to 1 August 2013 for the Annaberg-Buchholz site.

	Annaberg-Buchholz		
	Minimum PNC [cm^{-3}]	Mean PNC [cm^{-3}]	Maximum PNC [cm^{-3}]
10–20 nm	0	0.1	2.0
20–30 nm	0	0	1.0
30–50 nm	0	0.1	1.8
50–70 nm	0	0	1.4
70–100 nm	0	0	0.7
100–200 nm	0	0.1	1.0
200–800 nm	0	0	0.2

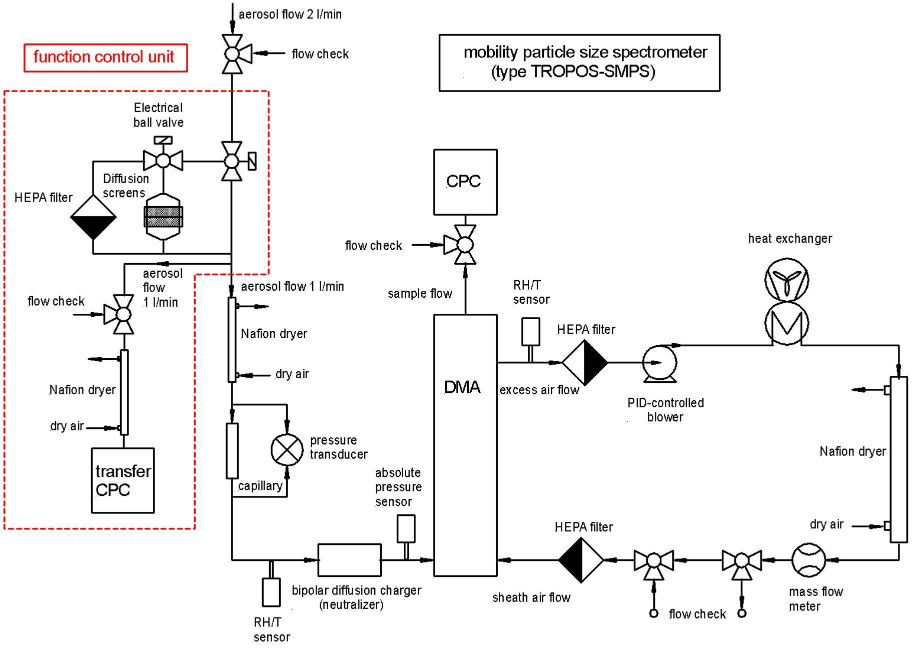

Fig. 1. Schematic sketch of a closed-loop mobility particle size spectrometer equipped with an optional function control unit.

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀I
▶I
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

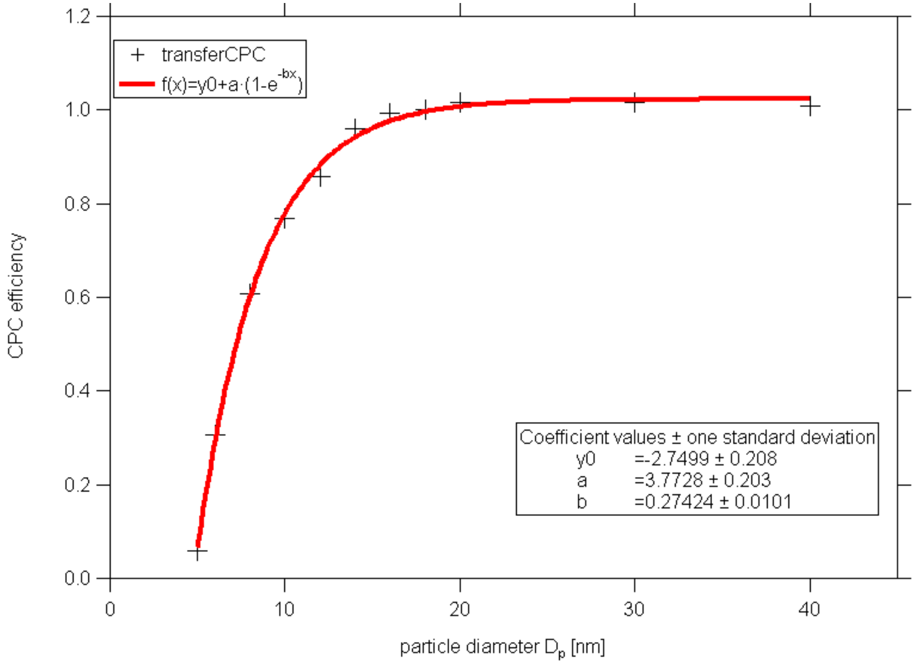

Fig. 2. Detection efficiency curve of the transfer CPC recorded on 24 October 2012.

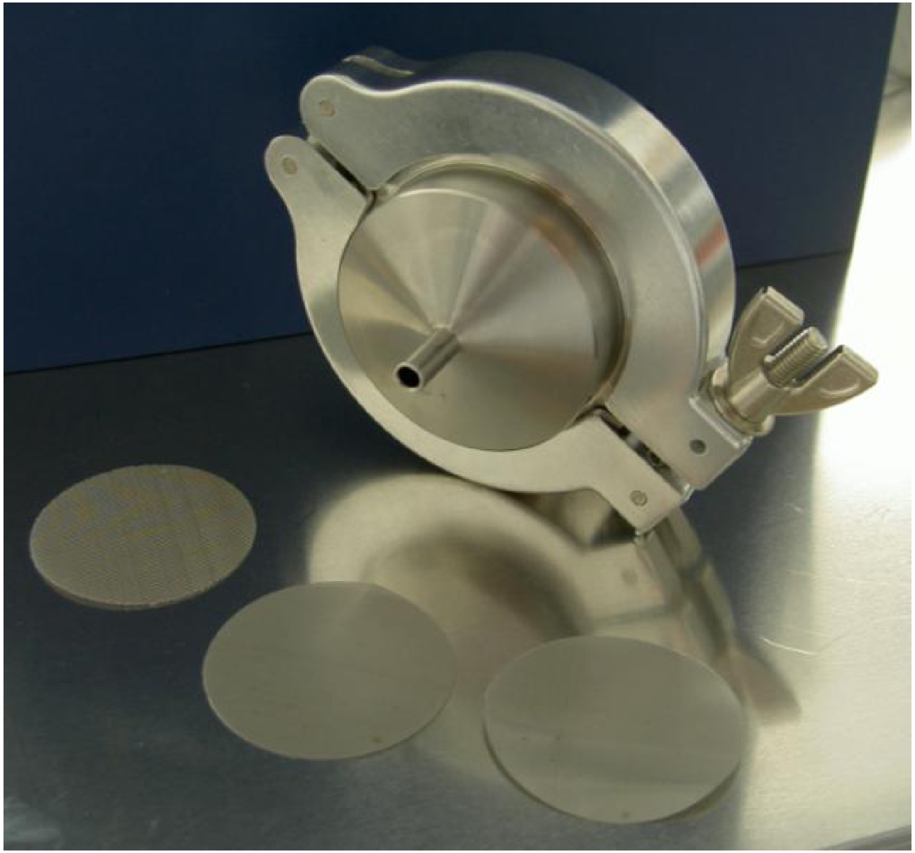

Fig. 3. Picture of diffusion screens with a stainless steel screen holder in the background.

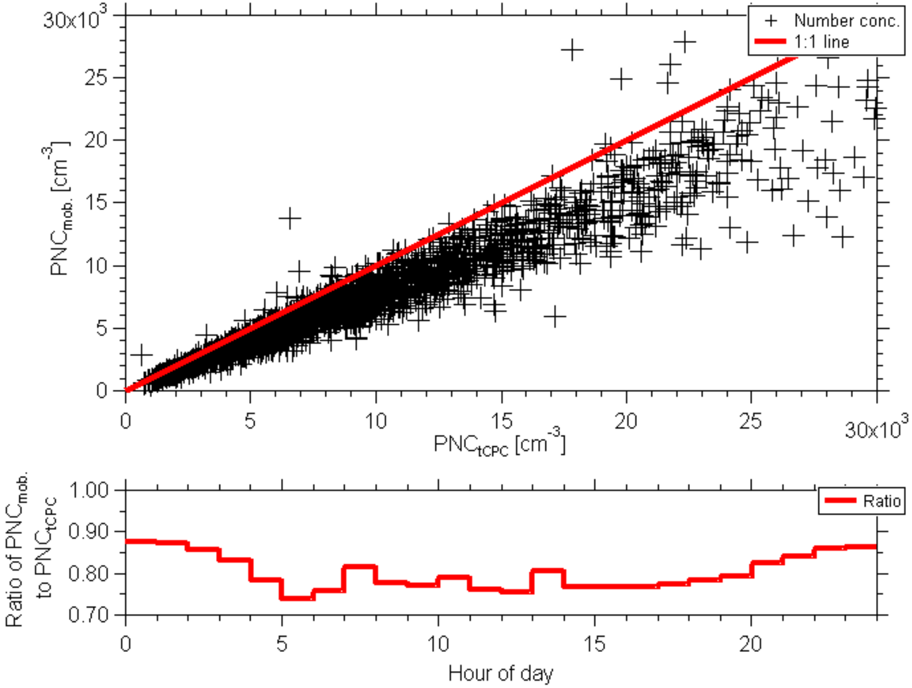

Fig. 4. Intercomparison of total PNCs from the mobility particle size spectrometer without diffusion screens and the transfer CPC for the Annaberg-Buchholz site from 1 August 2012 to 1 August 2013. Upper graph: scatter plot of $\mathrm{PNC}_{\mathrm{mob.}}$ against $\mathrm{PNC}_{\mathrm{tCPC}}$ (black crosses). Lower graph: diurnal variation of the ratio of $\mathrm{PNC}_{\mathrm{mob.}}$ to $\mathrm{PNC}_{\mathrm{tCPC}}$.

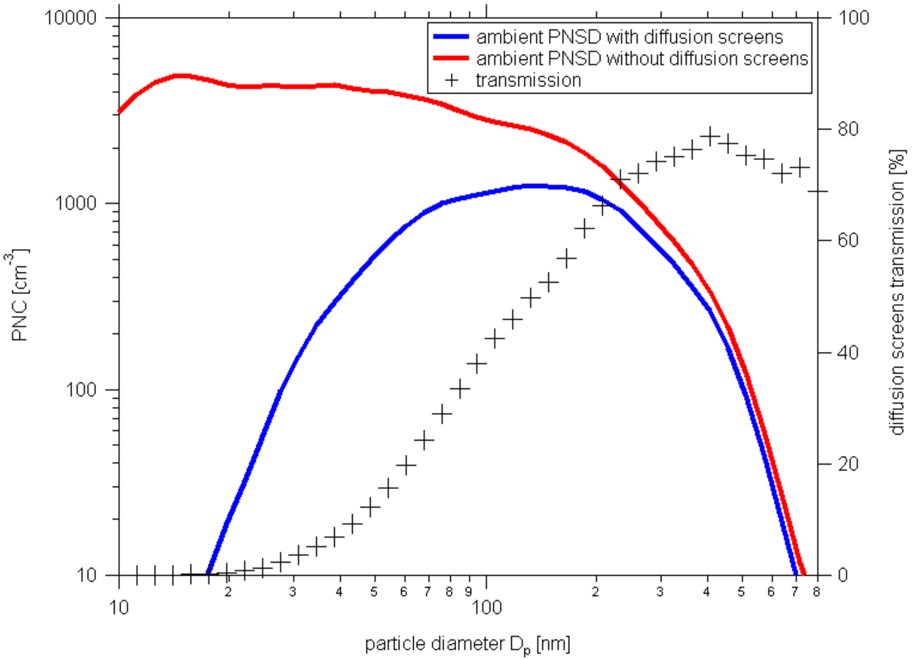

Fig. 5. Synchronous measurement of a PNSD with (blue solid line) and without (red solid line) diffusion screens. Black crosses represent the transmission of the diffusion screens.

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀❙
▶❙
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

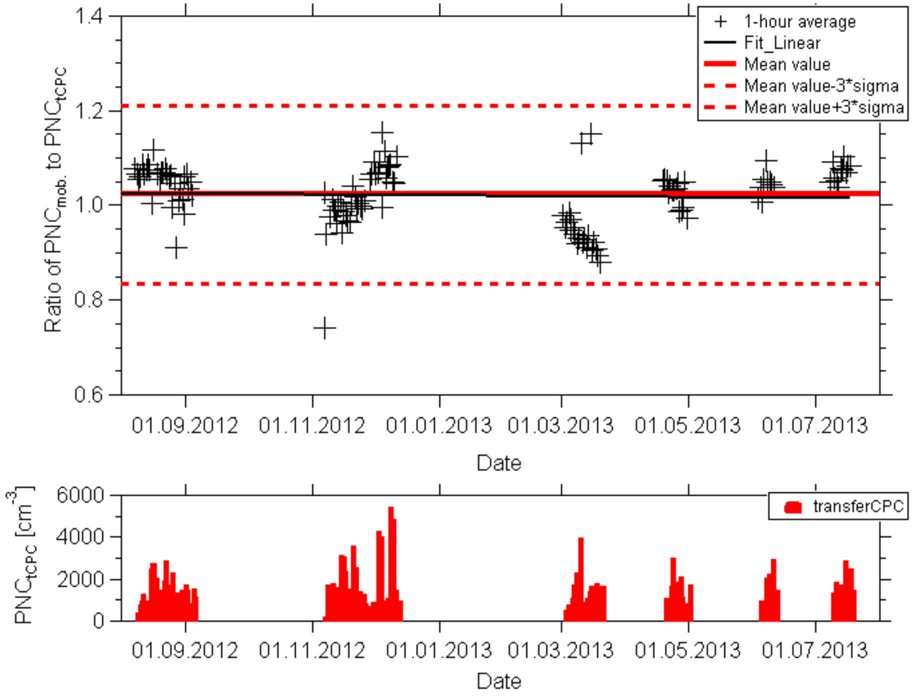

Fig. 6. Results from the diffusion screen measurements for the Annaberg-Buchholz site from 1 August 2012 to 1 August 2013. Upper graph: quality control chart shows entire mean value (red solid line), 99th confidence interval (red dashed line), and linear fit (black solid line) adapted from 1 h averages (black crosses). Lower graph: time series of the PNC from the transfer CPC.