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Design and performance characteristics of a condensation particle counter UF-02proto

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In this paper we present a design of a new swirling flow condensation particle counter UF-02proto, which has a continuous sampling flow. The performance and characteristics of the instrument were investigated. We studied experimentally the concentration range of the instrument and the detection efficiency as a function of particle diameter for silver particles. The lower cut-off size of the CPC, i.e. the limiting size when 50% of the particles are successfully accounted for, was determined to be 4.35–4.46 nm, depending slightly on a form of a fitted exponential step-function. The counting efficiency of the CPC at high particle concentrations was experimentally investigated using 20 nm silver particles. The maximum observable number concentration with a single particle counting method was approximately 100 000 cm^{-3} with an accuracy of 20%. The operation of the CPC UF-02proto was also compared with that of a commercially available CPC when operating both instruments in parallel. The CPC proved to be suitable for a variety of applications with a wide range of particle concentrations and particle sizes.

Introduction

The number concentration of aerosol particles has been measured using condensation particle counters (CPCs) for more than 100 years. These instruments can efficiently detect particles that cannot be detected by optical methods owing to the small particle size (McMurry 2000a). The working principle of the CPCs consists of three consecutive processes: supersaturating aerosol-laden air with vapour of a working fluid, growth of the particles by condensation of the supersaturated vapours and optical detection of the particles after their growth. The CPCs can be classified according to the technique that is used in creating the supersaturation. Currently, three main methods have been introduced: adiabatic expansion of an aerosol–vapour mixture (Metnieks and Pollak 1959), conductive cooling (Bricard *et al*. 1976, Sem 2002) and mixing

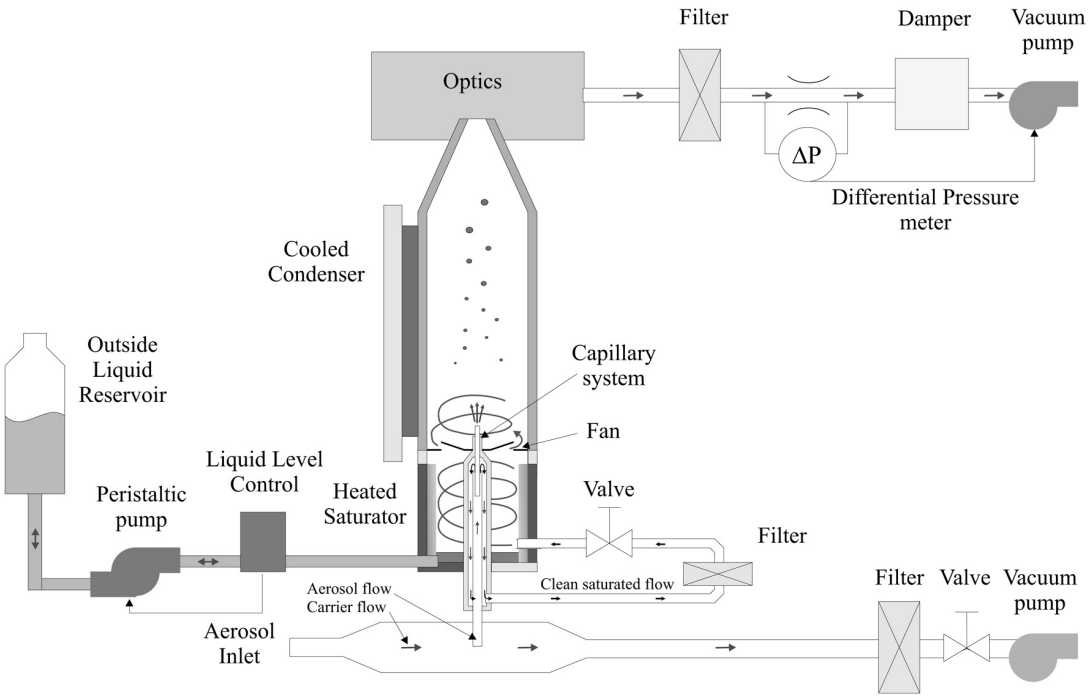

Fig. 1. A schematic illustration of a continuos flow condensation particle counter UF-02proto. The instrument uses a high flow rate (1.929 l min^{-1}) of the carrier. From the carrier flow, the aerosol flow (0.2746 l min^{-1}) is extracted by a capillary. This aerosol flow is divided into two. The first one (0.0322 l min^{-1}) is directed to the condenser. The second flow (0.2424 l min^{-1}) is circulated through a HEPA filter and a saturator block, in which the flow is saturated with respect to *n*-butanol and then mixed with the aerosol-laden air at a cooled condenser. This mixing generates a supersaturated region with respect to *n*-butanol. The butanol vapour condenses onto the aerosol particles within the sample flow and large droplets are formed, which are then focused to the optics and detected based on the scattering of light.

of cool and warm saturated air (Kousaka *et al*. 1982, Mavliev 2002, Wang *et al*. 2002).

A continuous flow condensation particle counter enables the counter to be used as a detector in applications that require a steady and incessant sampling flow. These applications often include differential mobility analysers and diffusion batteries as classifiers before the counter (McMurry 2000b). Another advantage of using a continuous flow model is the ability of CPCs to count individual particles. This enables measurements also at low number concentrations.

The properties of the vapour (and particles) and the degree of the supersaturation inside the CPC determine the lowest size of a particle on which the condensation can be initiated (e.g. Hämeri *et al*. 2002). In practical applications, many other factors (e.g. geometry, inlet design and flows) affect the true smallest detectable size (Stolzenburg and McMurry 1991). Because of these factors, all the CPC models have limitations in their operation. The main limitations concern the maximum concentration that can be reliably measured and the lowest detectable particle size. In practice, the largest detectable size is influenced strongly not only by the instrument characteristics but also by the whole instrument setup. However, the number concentration is typically dominated by the sub-micrometer particles and therefore the smallest detectable size is much more a significant parameter.

In this paper we present a design of a new CPC UF-02proto. The performance and characteristics of the instrument were investigated. A study of a detection efficiency as a function of particle size was conducted for silver particles. Also the reliable concentration range was investigated experimentally as well as the coincidence correction i.e. multiple particles falsely counted as one, was determined, and applied to

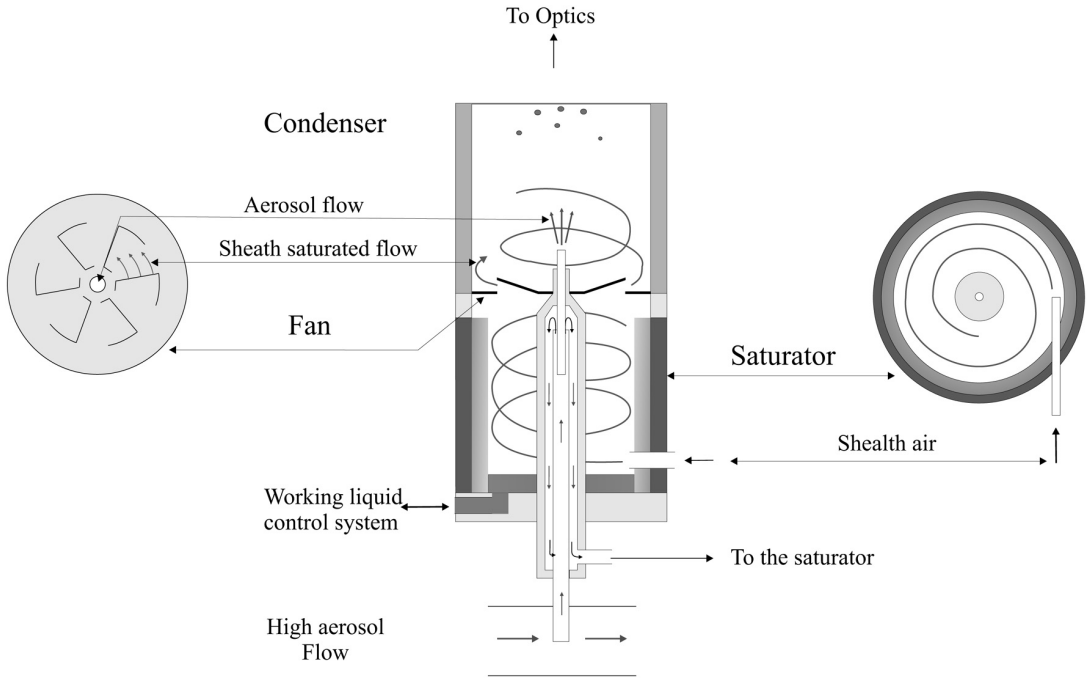

Fig. 2. A more detailed illustration of turbulent mixing inside the UF-02proto. The *n*-butanol saturated air swirls around the aerosol sample flow minimizing the diffusional losses to the walls of the condenser and maximizing the supersaturated region in which the particles are growing due to condensation of butanol vapour.

the hardware of the instrument. In addition, the CPC UF-02proto was operated in parallel with a commercial CPC (TSI Inc. model 3022) measuring laboratory air to assure the suitability of the instrument for ambient sampling.

Instrument design

A cornerstone of a continuous flow condensation particle counter design lies in a stable generation of supersaturation inside the apparatus and in well-established optics and electronics as well as in effective transport of the sampled aerosol and activated droplets within the counter. A schematic picture of a UF-02proto condensation particle counter is presented in Fig. 1. A more detailed illustration of a swirling flow generated inside the saturator-condenser is illustrated in Fig. 2. General operation parameters are listed in Table 1.

To minimize transport losses, the CPC UF-02proto uses a higher flow rate for sampling (a carrier flow) than what is used to detect the particles. From the carrier flow (1.929 l min^{-1}), an aerosol flow (0.2746 l min^{-1}) is extracted with a capillary tube. Furthermore, the aerosol flow is divided into two parts: one (0.2424 l min^{-1}) passing through a high performance filter and a saturator block and the other one (0.0322 l min^{-1}) directed to a growth tube. The values of

Table 1. General working characteristics of the CPC UF-02proto.

Temperature of saturator (°C)	42.97 ± 0.03
Temperature of condenser (°C)	10.45 ± 0.08
Temperature of optics (°C)	50 ± 1
Total flow rate (cm^3 min^{-1})	1929 ± 8 (0.4 %)
Aerosol flow rate (cm^3 min^{-1})	274.6 ± 1.6 (0.6%)
Flow through saturator (cm^3 min^{-1})	242.4 ± 0.8 (0.3%)
Mixing ratio	0.88
Pulse width (on comparator level) (μs)	2.5–3.0
Pulse amplitude (for 20 nm Ag, 10 000 cm^{-3}) (V)	2.04–5.00
Noise level (mV)	80
Comparator level (mV)	200
High voltage (V)	800
Wavelength (nm)	655
Thickness of laser beam (μm)	12
Laser power supply (V)	2.0
Power of laser beam by a calibration (mW)	2

the flow rates are a result of a trade-off between minimized transport losses due to diffusion and deteriorated counting statistics.

Inside a saturator block, working fluid (*n*-butanol) is evaporated from a liquid pool to the passing air flow. The liquid level within the saturator is maintained with a peristaltic pump, which transfers *n*-butanol from an outside reservoir on demand. When the counter is switched off, the pump removes all the butanol from the pool to the reservoir. This protects the optics from flooding of liquid working fluid e.g. during a relocation of the UF-02proto counter.

The aerosol-laden sample flow and butanol-saturated flow are combined together inside a cooled condenser. The aerosol flow enters the condenser at centreline. The saturated sheath flow is directed around the aerosol flow (Fig. 2). In order to enhance the mixing of the saturated air and cooler aerosol flow, a fan is used. In this way, the loss of the smallest particles to the walls of a condenser is minimized and, additionally, the supersaturated region is enlarged compared to a laminar-flow situation. In the condenser, high supersaturation levels with respect to butanol are obtained due to this efficient mixing between the cold sample and heated saturator flows. In the supersaturated region, particles within the aerosol flow grow into droplets large enough to be detected by optical methods. A conical shape of the condenser accelerates and focuses the particles to a measurement volume of an optical system.

The optical system of CPC UF-02proto consists of a laser, collection optics, absorbers, photomultiplier, analogue signal converter and microprocessor. The laser produces a light beam, which is focused to a measurement volume by the collection optics. As each activated droplet passes through the laser beam, it scatters light in all directions. A portion of light is captured by the photomultiplier. The remaining part is absorbed inside the optics. The photomultiplier converts the collected light pulse into an electrical signal that is then converted into a digital signal and counted by the microprocessor. The particle number concentration is calculated from the counted electrical pulses using a live-time counting method (e.g. Sem 2002). The counting time is user-selectable. The lower limit is one second.

Calibration apparatus and experimental setup

A design of a CPC determines the performance characteristics of the device. The two most important properties of a CPC are the particle detection efficiency as a function of particle size and maximum detectable number concentration. Scheibel and Porstendörfer (1983) described a measurement method to determine the particle detection efficiency. The same method was used also to examine the particle detection efficiency of the CPC UF-02proto in this work. The experimental setup of the measurements is presented in Fig. 3 (*see* also Aalto *et al*. 2001).

A tube furnace (Carbolite Furnaces MTF 12/388) was used to produce a polydisperse silver aerosol. At approximately 1000 °C, silver was evaporated into 3.5 l min^{-1} nitrogen flow. As the flow cooled down outside of the heated section of a tube furnace, the metal vapour nucleated and formed polydisperse silver particles. The aerosol particles were charged with an alpha-active ^{241}Am bipolar source. Then, a monodisperse fraction was selected with a differential mobility analyser (DMA VIE-08, Hauke, length 0.109 m). The DMA was operated with 2.0 l min^{-1} aerosol and 20.0 l min^{-1} sheath and excess air flows with an open loop arrangement. The classified 2.0 l min^{-1} aerosol was diluted with 3.6 l min^{-1} dry clean air to reduce the aerosol concentration below 10^4 cm^{-3}. This ensured that a coincidence inside the UF-02proto counter did not exceed 2%. For high aerosol particle concentrations, many particles are within the measurement volume of the laser at a same time. As a consequence, the optical signal from these particles cannot be distinguished and these particles are falsely counted as one.

The monodisperse, diluted sample was divided to the CPC UF-02proto and to an aerosol electrometer (TSI Inc. model 3068). The sample flow rates were 1.9 l min^{-1} and 3.7 l min^{-1} for UF-02proto and the electrometer, respectively. All flows were measured with a bubble flow meter (Gilian Gilibrator2) and maintained with needle valves.

The concentration signal from the UF-02proto was compared with the reference electrometer concentration reading. The detection efficiency

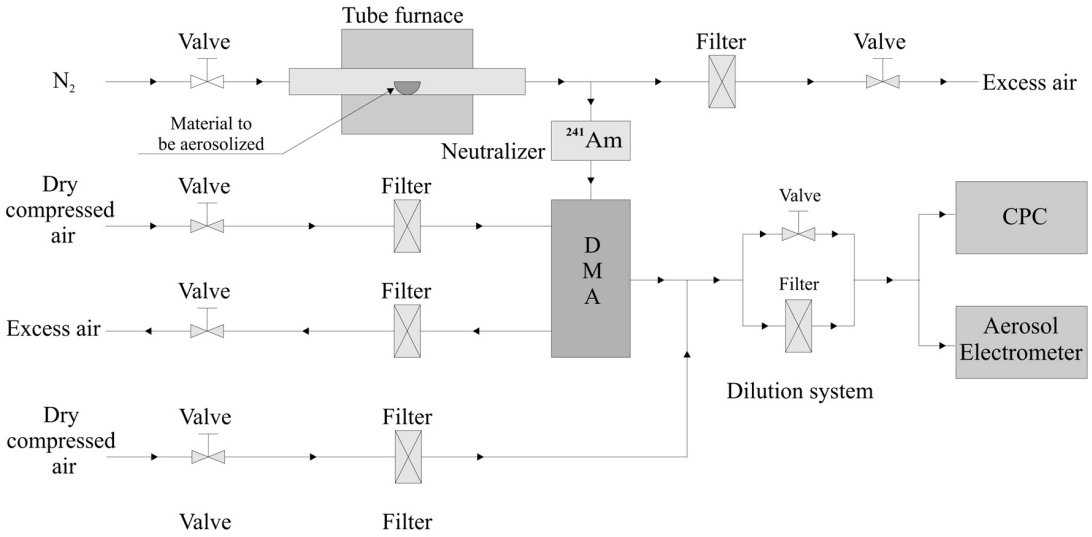

Fig. 3. Experimental setup for determining particle detection efficiency and concentration range of UF-02proto condensation particle counter. Silver was evaporated into nitrogen flow. Subsequently it cooled and formed ultrafine aerosol particles, which were charged (^{241}Am-source) and size segregated with a differential mobility analyzer (DMA). The sample was diluted in both the detection efficiency and concentration range experiments. A diluted monodisperse aerosol sample was divided to UF-02proto condensation particle counter and a reference electrometer.

of the CPC UF-02proto was determined as a ratio of the two measured concentrations. Since the generated particles in this study were smaller than 30 nm in diameter, they were most probably no more than singly charged (Wiedensohler 1988). This justified the use of an electrometer as a reference instrument.

In experiments to test the concentration range of the UF-02proto, monodisperse 20 nm silver particles were generated with variable concentrations. The absolute minimum concentration, i.e. background count of the UF-02proto, on the other hand, was specified by placing a high performance filter in front of the counter.

The optical system of the CPC UF-02proto was investigated separately at high concentrations of 20 nm silver particles. In the optical system the light scattered by the particles is focused to a photomultiplier. The photomultiplier converts the collected light pulses to electrical pulses. During the test, the photomultiplier voltage was set to 800 V. After amplification, the electrical pulse amplitude and background as well as pulse width were investigated using an oscilloscope. At a high particle concentration, pulse width increased due to coincidence of the electrical pulses. This is a result of multiple aerosol particles present inside the laser beam simultaneously. A coincidence correction to the measured concentrations was determined and applied.

The detection efficiency of the UF-02proto as a function of particle diameter was also investigated with the setup presented in Fig. 3. For this purpose, silver particles of variable size from 3 nm to 12 nm in diameter were generated. The silver particle size distribution generated with the tube furnace was unimodal with a peak value at 6 nm in diameter (Fig. 4). In order to eliminate the influence of a variable concentration in a detection efficiency experiment, a dilution system was applied. The aerosol sample was diluted to a constant concentration of 2500 cm^{-3} in each particle size.

Results and discussion

Working characteristics

The performance of a CPC in a long run depends crucially on the design of the instrument. The reliability of particle detection rests on the stability of the temperature gradients established

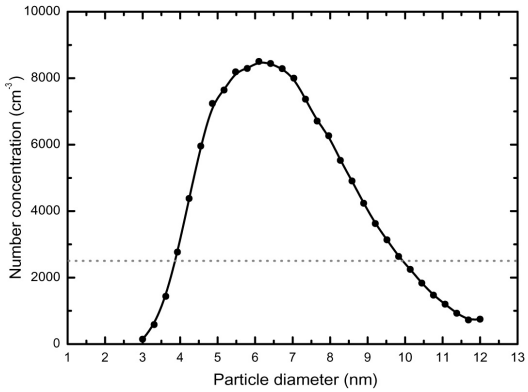

Fig. 4. A sample of oven-generated unimodal silver particles as seen by the reference electrometer. For detection efficiency studies, a dilution system was applied so that a constant concentration was obtained (dotted line).

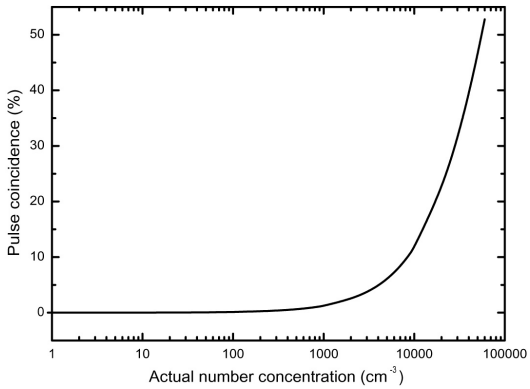

Fig. 5. A calculated coincidence of electrical pulses as a function of an actual number concentration measured with a reference electrometer. The exponential growth of the coincidence was applied to the hardware of the instrument. This extended the concentration range of the UF-02proto up to 100 000 cm^{-3}.

inside the CPC. Furthermore, the optical system counting the particles as well as the attached electronics should be produced with great care in order to successfully observe particles with a large enough concentration range. In addition, the flow system has to deliver the aerosol sample first to a supersaturated region, where the particles activate and then the activated droplets have to be transported efficiently to the optical detection unit. During the transport the particle losses have to be minimised and controlled.

The working characteristics of an UF-02proto swirling flow condensation particle counter are presented in Table 1. During a one-hour measurement, the temperatures of the walls of the saturator and condenser were monitored with built-in temperature sensors (Dallas, DS1626). The measured temperatures were 10.45 ± 0.08 °C and 42.97 ± 0.03 °C for the condenser and the saturator, respectively. Fluctuations in both temperatures were smaller than 1.0%. The temperature of the optics (50 ± 1 °C) had the strongest variation of 2% due to the fact that the pulsed laser generated a fluctuating heat load to the system. The stable temperatures inside the CPC provided a well-controlled supersaturation inside the condenser. This enabled a reliable and repeatable growth of the sampled particles, which were introduced to the counting optics with a steady flow rate.

The optical system of the CPC UF-02proto was investigated separately using a variable concentration of monodisperse 20-nm-diameter silver particles. At a high particle concentration, the pulse width increased due to coincidence of the electrical pulses. This was a result of multiple particles present at a measurement volume of a laser beam. The particles were incorrectly counted as one. The coincidence of UF-02proto at a high concentration of aerosol particles as a function of a concentration measured by the reference electrometer is illustrated in Fig. 5. For a concentration of 1000 cm^{-3}, the pulse coincidence was smaller than 1%. However, at 10 000 cm^{-3} it was already 11.75%. It was higher than 50% for concentrations larger than 50 000 cm^{-3}. Thus, the coincidence increased exponentially as a function of aerosol number concentration after an aerosol particle concentration of 10 000 cm^{-3}. To eliminate this problem, a live-time counting method (Sem 2002) was included in the concentration calculation algorithm. This broadened the concentration measurement range up to 100 000 cm^{-3}. The algorithm was installed in the microprocessor of the CPC and it was used to calculate particle number concentration at all concentration ranges.

Concentration range

A minimum detectable number concentration is determined by false background counts in the optics. For these investigations, a HEPA filter

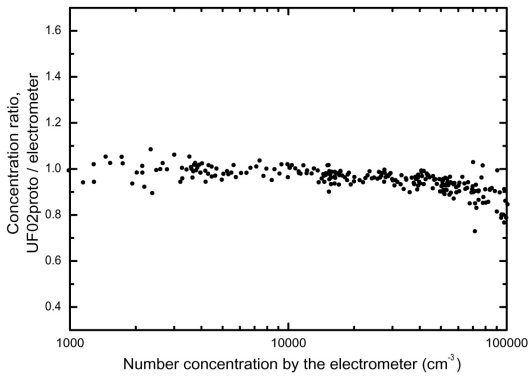

Fig. 6. A ratio between the measured concentrations of UF-02proto condensation particle counter and the electrometer as a function of electrometer concentration. The scatter at the lower end is a result of poor counting statistics of the electrometer at these concentrations. At the high end, the counting efficiency of the particle counter deteriorated.

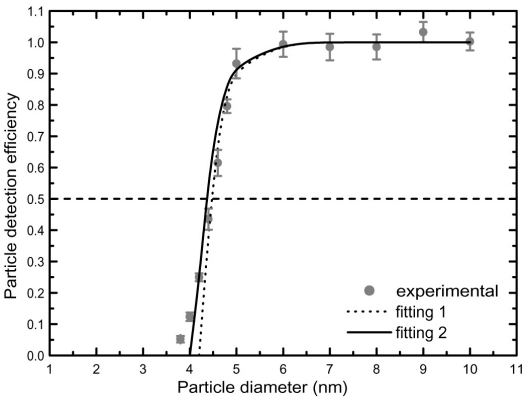

Fig. 7. Particle detection efficiency of a UF-02proto as a function of the particle size for silver particles. Two fittings give almost identical values for the cut-off size of 4.35–4.46 nm.

was connected into the inlet of the CPC. The measurements of the concentrations were conducted during one hour. The average number concentration during the measurement period was calculated. The results show that the false background was only 0.002 cm^{-3}, when the temperature difference between the saturator and the condenser was set to 32.5 °C. In principle, the existence of this background can be explained by particle production due to homogeneous or ion-induced nucleation inside the CPC or by the noise of the electronics. However, as shown recently by Kulmala *et al*. (2005) homogeneous nucleation is improbable under these conditions.

The maximum measured number concentration of the CPC UF-02proto was investigated comparing the number concentration measured by the CPC with that of the reference electrometer. The experiments were performed using a monodisperse aerosol with a diameter of 20 nm (Fig. 6). Relatively large fluctuations in the ratio between the concentrations of the two instruments were observed for concentrations < 3000 cm^{-3} and the best correlation was in the concentration range 3000–50 000 cm^{-3}. For particle concentrations > 50 000 cm^{-3}, the measured concentration by the CPC UF-02proto was smaller than the concentration measured by the electrometer. The difference was higher than 20% for concentrations > 100 000 cm^{-3}. At higher particle concentrations, the coincidence increased exponentially as a function of particle concentration and the small errors in the coincidence correction start to add up. This results in a decrease in the measurement accuracy at the highest concentrations. Thus, the device cannot determine aerosol number concentration larger than 100 000 cm^{-3} accurately (±20%).

The investigations of the measured concentration range showed that the CPC can be used to measure the particle number concentration from 0.002 up to 100 000 cm^{-3}. The summary of the concentration range experiments is presented in Table 2.

Detection efficiency

One of the main characteristics of a CPC is the particle detection efficiency E as a function of particle diameter d. As a measure of detection efficiency, a ratio between the concentration measured by UF-02proto and that measured by the reference electrometer as a function of particle diameter is chosen (Fig. 7). The detection

Table 2. Concentration range and accuracy of the CPC UF-02proto.

False background counts (particles cm^{-3})	0.002
Maximum number concentration accuracy ± 10% (particles cm^{-3})	50 000
Maximum number concentration accuracy ± 20% (particles cm^{-3})	100 000

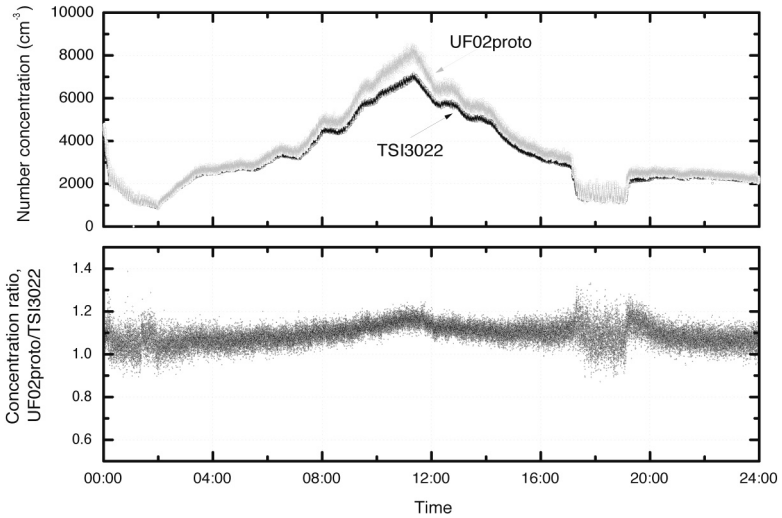

Fig. 8. A time series of measured indoor air aerosol particle number concentrations by the CPC UF-02proto and CPC TSI model 3022 during one day (upper panel), as well as the concentration ratio between the two instruments (lower panel).

efficiency reached a value of 1 at large particle sizes and it was smaller than 0.9 for particles smaller than approximately 5 nm. The detection efficiency decreased rapidly and was smaller than 0.1 for particles smaller than 4 nm in diameter. The measured detection efficiency curve was very steep providing a well-defined smallest observable size.

In order to determine a cut-off size, i.e. the size when 50% of the particles were counted, two step-wise functions were fitted to the experimental data. The first function had a form of

$$E(d) = 1 - \frac{\lambda_1}{1 + e^{(d - \lambda_2)/\lambda_3}}, \quad (1)$$

where λ_1 = 5.0, λ_2 = 3.65 and λ_3 = 0.32 are parameters determined based on a least-squares fitting. According to the fitting, the cut-off size was 4.35 nm. Additionally, the cut-off size was determined with a function

$$E(d) = 1 - e^{(\alpha_1 - d)/\alpha_2}, \quad (2)$$

where α_1 = 4.25 and α_2 = 0.3 are the fitted parameters. As a result, the cut-off size of 4.46 nm was obtained.

The results of both fittings and experimental data are presented in Fig. 7. The second fitting had a better agreement with the experimental data for small particles. However, considering experimental uncertainties, both functions gave almost equal values for the cut-off diameter.

Ambient aerosol measurements

The CPC UF-02proto was tested by operating the instrument in parallel with another commercially available particle counter (TSI Inc. CPC model 3022, Sem 2002). Both CPCs were sampling laboratory air. The working fluid of both was *n*-butanol. The averaging time was one second. A comparison of the measured concentrations by the two instruments is shown in Fig. 8. During the experiment, the instruments detected particle concentrations between 1000 and 9000 cm^{-3}. The two CPCs agreed rather well. However, the CPC TSI3022 measured slightly lower values. The relative difference was typically 5% to 10%. A plausible explanation for this is the influence of different cut-off sizes of the instruments. The reported cut-off diameter of the CPC TSI3022 is about 7 nm (Quant *et al.* 1992), while that of the CPC UF-02proto, based on this study, is approximately 4.5 nm. In case of a large contribution of sub-10 nm particles to the total number concentration, the small differences in cut-off sizes lead to observed discrepancies between the number concentrations.

When comparing the presented CPC UF-02proto with other commercially available counters (e.g. TSI Inc. models 3010, 3022 and 3025), one has to bear in mind the purpose of the measurements. Two main features need then to be considered: the counting efficiency as a function of particle diameter and concentration range.

Not a single counter can be the most exclusive for all the possible applications and demands. If it is crucial to determine particle concentrations of sub-10 nm particles, UF-02proto compares favourably with TSI Inc. model 3010 (cut-off at 10 nm, Mertes *et al*. 1995) and TSI 3022 (cut-off at 7 nm, Quant *et al*. 1992). On the other hand, a counter designed to detect ultrafine particles (TSI Inc. 3025) has a lower cut-off size at 3 nm according to Kesten *et al*. (1991) than UF-02proto (4.5 nm, this study). In conditions of high particle concentrations, the maximum accurately observable concentration is the most important factor determining a suitable counter. The aforementioned counters have upper limits of 10 000 cm^{-3} (Mertes 1995), 10 000 000 cm^{-3} (Quant *et al*. 1992) and 100 000 cm^{-3} (Stolzenburg *et al*. 1991) for TSI models 3010, 3022 and 3025, respectively. The maximum accurately determined concentration with the UF-02proto is 100 000 cm^{-3}, which makes it comparable to a TSI 3025, when both instruments are operating in a single particle counting mode. Larger maximum concentration with a TSI model 3022 is a result of a photometric mode, where the number concentration is detected as a total attenuation of the laser beam due to particle population within the measurement volume.

Conclusions

A condensation particle counter UF-02proto was designed to detect ultrafine aerosol particles of few nanometres with high detection efficiency. The new design of the instrument is based on the swirling flow generated inside the saturator-condenser. The main parameters affecting the performance of the particle counter characteristics (temperature of the saturator, the temperature of the condenser, operating flow rates and the electronic parameters of the optical system) were studied. In addition, the particle detection efficiency as a function of the size was experimentally investigated in 3–10 nm particle diameter range. The cut-off size was determined by fitting an empirical function to the experimental data. The obtained value was 4.35–4.46 nm depending to a small extent on a form of a fitted exponential function. This is comparable to other commercial CPCs designed to detect ultra-fine particles efficiently. Also coincidence, resulting from multiple particles within the measurement volume of the optical system, was examined. A relevant correction was introduced to the hardware of the counter. The counting efficiency of the CPC at high particle concentrations was experimentally investigated using 20 nm silver particles. The maximum observable number concentration (accuracy 20%) was 100 000 cm^{-3}.

The UF-02proto condensation particle counter was shown to be suitable for a variety of applications with a wide range of particle concentrations and sizes. The ambient data obtained measuring laboratory air showed 5%–10% differences when compared with a TSI Inc. CPC 3022. We ascribe these differences to technical differences of the CPC models resulting in different cut-off sizes of the two instruments, which have to be taken into consideration when using any condensation particle counter.

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