

The standard operating procedure for Airmodus Particle Size Magnifier and nano-Condensation Nucleus Counter

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ABSTRACT

Measurements of aerosol particles and clusters smaller than 3 nm in diameter are performed by many groups in order to detect recently formed or emitted nanoparticles and for studying the formation and early growth processes of aerosol particles. The Airmodus nano-Condensation Nucleus Counter (nCNC), consisting of a Particle Size Magnifier (PSM) and a Condensation Particle Counter (CPC) is a versatile tool to detect aerosol particles and clusters as small as ca. 1 nm in mobility diameter. It offers several different operation modes: fixed mode to measure the total particle number concentration with a fixed, but adjustable lower cut-off size and stepping and scanning modes for retrieving size-resolved information of ca. 1–4 nm particles. The size analysis is based on changing the supersaturation of the working fluid (diethylene glycol) inside the instrument, which changes the lowest detectable size. Here we present a standard operating procedure (SOP) for setting up, calibrating and operating the instrument for atmospheric field measurements. We will also present recommendations for data monitoring and analysis, and discuss some of the uncertainties related to the measurements. This procedure is the first step in harmonizing the use of the PSM/nCNC for atmospheric field measurements of sub-3 nm clusters and particles.

1. Introduction

The Airmodus Particle Size Magnifier (PSM) was developed originally at the University of Helsinki to measure particles smaller

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than 3 nm in diameter in order to study atmospheric new particle formation and related processes. The prototype instrument is described [Vanhanen et al. \(2011\)](#). The instrument was soon thereafter commercialized by Airmodus Ltd. The first PSMs by Airmodus consisted solely of the magnifier part, where particles are grown. An external condensation particle counter (CPC) from another manufacturer was used to count the particles. Later, a nano-Condensation Nucleus Counter (nCNC, Airmodus model A11) was commercialized, which consists of the PSM, an Airmodus CPC (model A20) and a control software to run both instruments together.

The name “Particle Size Magnifier” originated from the work of [Okuyama et al. \(1984\)](#), who constructed a system for growing sub-10 nm nanoparticles. The term “PSM” is very often used synonymously to describe the whole PSM-CPC combination, although it strictly refers only to the pre-conditioner part. Here we mainly describe the use of the Airmodus A11 nCNC, although most of the principles hold also if the Airmodus PSM is operated together with any other CPC. We will also discuss general issues relevant for the measurement of sub-3 nm aerosol particles, which are common for all instruments measuring nanometer scale clusters and aerosol particles.

The PSM/nCNC has been used for measurements of the sub-3 nm particle concentration and size distributions at various locations, including the boreal forest ([Kontkanen et al., 2017](#); [Kulmala et al., 2013](#); [Sulo et al., 2021](#)), the Amazon ([Wimmer et al., 2018](#)), cities and polluted environments ([Kontkanen et al., 2017](#); [Yao et al., 2018](#); [Yu et al., 2013](#); [Zhou et al., 2020](#)), mountain tops ([Bianchi et al., 2016, 2020](#); [Rose et al., 2015](#)), Antarctica ([Jokinen et al., 2018](#)) as well as for determining nucleation rates and early growth rates during laboratory experiments (e.g. [Kirkby et al., 2016](#); [Lehtipalo et al., 2018](#); [Sipilä et al., 2010](#); [Yu et al., 2017](#)). In addition to atmospheric studies, it has been used to characterize nanoparticle concentrations originating from industrial processes ([Ahonen et al., 2017](#); [Sarnela et al., 2015](#)), traffic and engines ([Alanen et al., 2015](#); [Ronkko et al., 2017](#)) and 3D-printers ([Mendes et al., 2017](#)). Several technical studies or laboratory characterizations have explored its performance compared to reference instruments in different conditions and using different particle compositions ([Brilke et al., 2020](#); [Kangasluoma et al., 2014, 2015, 2016a](#); [Wimmer et al., 2013](#)). Additionally, methods for determining nucleation and growth rates using PSM data have been defined in several articles ([Dada et al., 2020](#); [Kulmala et al., 2012](#); [Lehtipalo et al., 2014](#)).

The aim of this protocol is to describe the best practices for operating the PSM/nCNC in field conditions and to unify the measurement procedures used by different groups, especially within the ACTRIS (Aerosols, Clouds and Trace gases Research Infrastructure) community, for achieving comparable, high-quality observations of sub-3 nm particles. In addition, we describe possible issues e.g. due to varying external conditions, which the users should be aware of. This protocol is based on almost 10 years of experience working with the Particle Size Magnifier at the University of Helsinki, as well as discussions between the users and instrument manufacturer. The procedures presented here are related to the current PSM/nCNC versions in use (A09/A11) and should be updated or revised upon significant technical development of the instrumentation.

2. Description of instrumentation and setup

2.1. Physical basis

The idea behind all CPCs is to create a supersaturation of the working fluid and grow the sampled aerosol particles by condensation to large enough sizes that they can be detected by optically. Different CPC types differ from each other mainly by how the supersaturation is created and by their working fluid. A historical review of different CPCs is written by [McMurry \(2000\)](#), while [Kangasluoma and Attoui \(2019\)](#) have recently reviewed the development of sub-3 nm CPCs and their calibration methods.

The nCNC/PSM, like any CPC, measures all airborne particles that grow (= those which “activate”) inside the instrument and are finally detected by the counting optics of the CPC, *i.e.* which act as condensation nuclei at the supersaturation level inside the instrument. At the high supersaturations needed to activate sub-3 nm particles, some of the condensation nuclei might actually be large molecules, molecular clusters and cluster ions rather than actual aerosol particles with macroscopic properties (e.g. well-defined surface, diameter, density and uncountable number of molecules). To describe the activated objects, the term “nano condensation nuclei” (nano-CN) was proposed by a few studies ([Lehtipalo et al., 2011](#); [McMurry et al., 2011](#)), while other groups have adopted the term “nanocluster aerosol” (NCA; e.g. [Ronkko et al. \(2017\)](#)), although it is also common to speak of clusters or sub-3 nm particles. Here we call the nano-CN measured by the nCNC simply as “particles”.

At a certain supersaturation, which is controlled by the temperature and flow rate settings and the working fluid of the particle counter, the activation probability of a particle depends on its size. This is due to the Kelvin effect, which tells that the smaller the particle, the higher the supersaturation needed for its activation. Therefore, at a given supersaturation, particles smaller than a certain diameter cannot in principle be counted. The lowest diameter at which the instrument still counts half of the particles is called the “cut-off diameter”, and the detection efficiency as a function of particle size is called the “cut-off curve”. Ideally, the cut-off curve is sharp or even a step-function (the instruments would count all particles larger than a certain size and none of the particles lower than that size). In practice, all the particles sampled by the instrument do not experience exactly the same supersaturation profile, and some might be lost due to diffusion during the transport inside the instrument, which affects the shape of the cut-off curve, making it less steep.

Another factor affecting the cut-off size of the instrument is the composition of the particles, due to vapor-seed interactions affecting the heterogeneous nucleation probability. For example, it is well known, that when water is used as a working fluid, hygroscopic particles have a lower detection limit than hydrophobic particles. Large deviations in the cut-off size have been reported especially for water-CPCs ([Kulmala et al., 2007](#); [Kupc et al., 2013](#)) and diethylene glycol CPCs ([Kangasluoma et al., 2014](#)), but also for n-butanol based CPCs ([Giechaskiel et al., 2011](#)). Therefore, since the ambient aerosol population can be externally mixed (consisting of particles with different chemical composition), and the aerosol composition especially in the nucleation mode is poorly known, this causes considerable uncertainty on the real cut-off size of the instrument, although it would be calibrated in the laboratory using a

certain of test aerosol (Dada et al., 2020). This also means that particles of the same size, but different chemical composition can have a different detection efficiency close to the cut-off size. This feature has been used to get indirect information about the composition of nucleation mode aerosol using a CPC-battery (Kulmala et al., 2007; Riipinen et al., 2009), but it adds to the uncertainty on the measured total concentration. In environments with a large number of nucleation mode aerosol (e.g. close to traffic or during new particle formation events), the total number concentration measured by a CPC depends strongly on its cut-off size (Kangasluoma & Kontkanen, 2017), as a big fraction of the total particle number concentration can be in the sub-10 nm range (Kontkanen et al., 2017; Zhou et al., 2020).

2.2. PSM

The Airmodus PSM, like all previous PSMs, is based on the principle of mixing of two flows in order to achieve supersaturation of the working fluid: the sample flow and the flow saturated with a vapor. The main advances of the Airmodus PSM, compared to the previous PSMs, are the engineering designs allowing continuous operation, also outside laboratory conditions.

The Airmodus PSM operates with diethylene glycol (DEG), which has been shown to be one of the most suitable working fluids for sub-3 nm particle activation (Iida et al., 2009). To minimize particle losses in the line between the inlet and the mixing region (Fig 1), the inlet flow rate of the PSM is 2.5 lpm (liters per minute), which is a higher flow rate than in most CPCs. The flow rates of the PSM are controlled with mass flow controllers (MFCs) and the flow rates are converted to volumetric flow rates assuming 20 °C and 101.3 kPa.

Downstream of the inlet, the sample flow is turbulently mixed with a heated flow coming from the saturator. The mixing piece is made of conductive plastic and it is not temperature controlled and thus the temperature of the mixed flow is a temperature weighted mean of the inlet and saturated flow rates.

The PSM saturator is a sintered metal tube, which is heated up to 70–85 °C. After the saturator has been wetted for the first time, DEG flows to the saturator passively via hydrostatic pressure. In the saturator, DEG evaporates into the saturator flow, which is usually filtered, compressed air. The saturator flow can be changed between 0.1 and 1.3 lpm (0.1 and 1 lpm in the older PSM models) to change the cut-off size of the instrument in the range of about 1–4 nm depending on temperature settings.

Downstream of the mixing section, there is a short conical section followed by a cylindrical growth tube. The growth tube wall temperature is typically set to 1–5 °C. In the Airmodus PSM, the largest DEG supersaturation along the particle trajectories is usually encountered in the growth tube, where particle activation and growth take place (Kangasluoma et al., 2016a). At the end of the growth tube, particles are sampled to a CPC from the centerline of the flow, typically with an inlet flow rate of 1.0 lpm, and the rest of the flow (~1.6–2.8 lpm) is discarded as excess flow. According to measurements by Vanhanen et al. (2011), the particle size after the growth tube is ca. 90 nm. A schematic of the main parts of the instrument is shown as Fig. 1.

2.3. CPC

As the particles activated and grown in the PSM are still not large enough to be counted optically, a CPC is needed after the PSM to grow the particles further and count them. In principle, any CPC can be used, but it is preferable that the CPC cut-off size is large enough so that the PSM and CPC cut-off curves do not overlap. Therefore, a cut-off size larger than >7 nm is recommended for the CPC. Also, it should be noted that the CPC determines the upper concentration limit of the PSM-CPC system, i.e. how large concentrations can be detected reliably, so this might affect the choice of the CPC.

In the Airmodus nCNC system, an A20 CPC by Airmodus is placed downstream of the PSM to count the grown particles (Fig. 1). The A20 is a typical laminar flow diffusion cooling-type CPC operated with butanol. Its inlet flow rate is 1 lpm, which is controlled with a critical orifice. The sample flow passes through a heated saturator in which butanol saturates the flow. Butanol condensation and particle growth to optical sizes take place in the condenser. The temperature settings of A20, when used together with a PSM, are chosen so that the cut-off is around 10 nm (saturator 35 °C and condenser 20 °C) to avoid any overlap in the PSM and CPC cut-off curves.

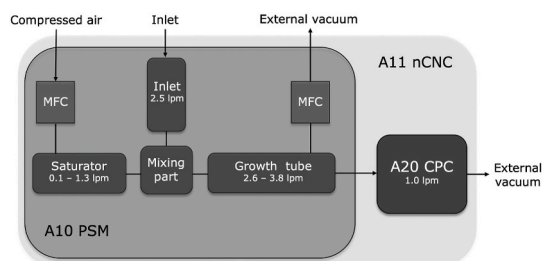


Fig. 1. Schematic of the A11 nano Condensation Nucleus Counter showing the main parts of the PSM, the CPC and the flow rates through each part (adapted from the A11 user manual). MFC = mass flow controller.

2.4. Measurement setup

The installation process and correct setup of the nCNC/PSM are explained in the instrument manual. However, there are some things to consider regarding placing of the instruments, sampling and environmental conditions, which require attention from the user. We summarize these best practices regarding the ambient measurement in the sections below.

2.4.1. Placing of the instrument

The outlet of the PSM should be connected to the inlet of the CPC with conductive tubing, typically the 45 cm conductive silicon tube provided with the instrument (the instrument is calibrated by the manufacturer using this tube). Using a shorter or longer tube is possible, but it can affect the response time of the instrument. The CPC should be placed on a higher level than the PSM outlet, e.g. on top of the PSM (see Fig. 2A). This is because small amounts of DEG could exit the PSM outlet and condense into the tube between the PSM and CPC. If the CPC is below the PSM, the condensed DEG flows directly into the CPC, where it might contaminate the saturator and the optics. A small drip bottle or T-piece before the connecting tube can be used to collect the excess DEG (Fig. 2B).

It is important that the DEG fill bottle is placed directly on top of the PSM, as DEG flows from the fill bottle to the saturator with hydrostatic pressure. Placing the fill bottle higher than that may unnecessarily increase DEG consumption, increasing also the need to drain the DEG reservoir. If the bottle is lower than the PSM, DEG does not flow into the saturator. Fig. 2 shows the correct setup of the instrument.

During installation, one needs to pay attention that there is proper ventilation around the PSM. The instrument takes air from the back of the instrument and it flows out below the instrument. If the pathway for cooling air is interrupted or the temperature in the measurement room is high, the PSM's internal temperature may increase too much for it to operate properly. Typically, the first indicator of too high PSM enclosure temperature is a warning of the drain temperature. The next symptom is the instrument's inability to keep growth tube temperature at the set point value, which will affect the detection efficiency of the instrument. We recommend keeping the instrument inside a temperature-controlled measurement site/building.

2.4.2. Inlet recommendations

In any measurements of sub-10 nm particles, the length of sampling lines must be minimized to avoid unnecessary particle transport losses. Sampling with traditional aerosol inlets, such as vertically through the roof with a PM2.5 impactor inlet, can cause significant particle transport losses in the sub 10 nm range.

To minimize particle losses in the sampling lines, there are few general considerations: 1) the sampling lines should be as short as possible, 2) the flow should be laminar and 3) all unnecessary bends, valves and changes in the tube inner diameter should be avoided. This leads to the conclusion that the sampling line should be a straight tube with laminar flow. If the sampling line is anything else, the particle transmission should be experimentally characterized down to the sizes of interest. In practice, the shortest possible sampling line length is often achieved by drilling a hole through the wall and placing the PSM inlet horizontally as close as possible to the wall.

The transport losses can be further minimized by using a core sampling configuration, in which, at the end of the sampling line, a portion of the flow is subsampled to the instrument from the flow centerline. This allows using a higher transport flow in the main inlet and then sampling the highest particle concentration in the flow profile (Fu et al., 2019; Kangasluoma et al., 2016a).

Fig. 3 shows an inlet system that was developed at the University of Helsinki for the atmospheric measurements with the PSM, and is described in more detail by Kangasluoma et al. (2016a). The purpose of the inlet system is to minimize particle diffusion losses during atmospheric sampling, and to allow regular automatic monitoring of the background counts of the PSM by providing particle free atmospheric air to the PSM at regular intervals (see chapter 3.2.3 for details of background measurements). Minimization of the diffusion losses is achieved through the core sampling method in which particles are extracted from the center of a main sampling flow

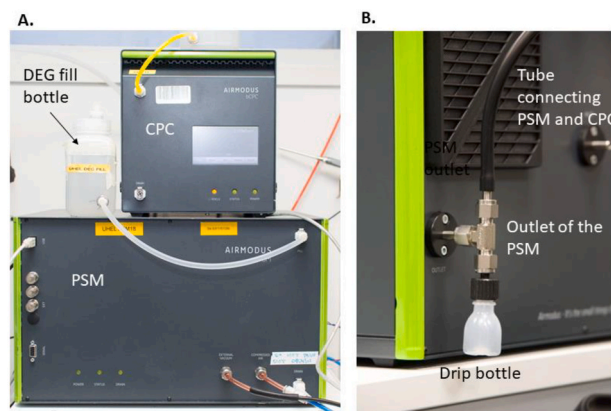


Fig. 2. Example of a correct setup of the instrument. **A.** The DEG fill bottle for the PSM is placed directly on top of the PSM and the CPC is placed on a higher level than the PSM. **B.** The PSM outlet and CPC inlet are connected to each other with conductive tubing and a drip bottle is placed at the PSM outlet.

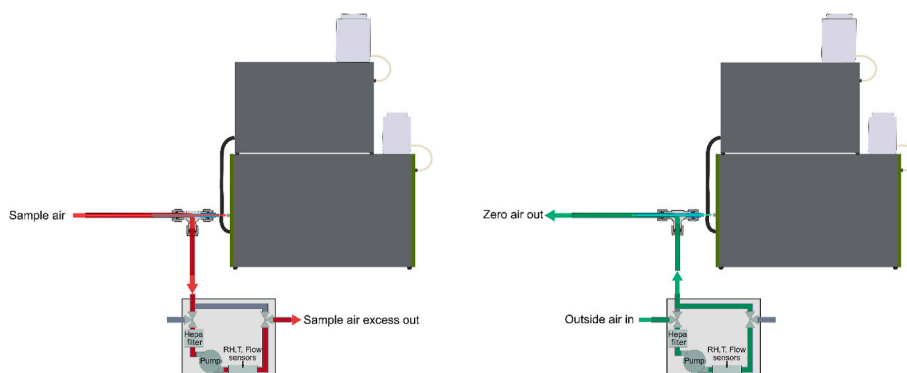


Fig. 3. The PSM setup with a core sampling inlet that can reverse the flow so that particle-free air is measured for monitoring the instrument background automatically. Red arrows show the inlet flow directions in normal operation mode and green arrows in background measurement mode. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

where their concentration is larger than at the outer edges. During atmospheric sampling, bypass flow is drawn with an external blower through the main inlet until the core sampling piece (Fig. 3, left side). For background measurements, the two 3-way valves are switched so that the blower draws air from the atmosphere through a particle filter, and leads that air through the core sampling piece to the PSM (Fig. 3, right side).

2.4.3. Consideration of environmental conditions

Depending on the location and conditions, drying or diluting the sample before measurement might be necessary. In such cases, the whole inlet system should be designed and characterized carefully, as sub-3 nm particles are very susceptible to losses. Depending on the composition, particles may also evaporate or grow hygroscopically if sample conditions (especially temperature and relative humidity) are changing during sampling.

For sampling ultrafine particles, the Global Atmospheric Watch program recommendation, followed also by ACTRIS, is to condition the sample to a RH lower than 40% (WMO, 2016). However, traditional aerosol driers (such as diffusion or membrane driers) are usually not suitable for sub-3 nm particles, as they introduce large losses for the smallest particles. There is also indication that water vapor increases the detection efficiency of DEG-based counters (Rörup et al., 2021), but more research is needed to fully understand and quantify the effect. Therefore, it is not yet known, how suitable the 40% RH limit is for the reliable measurement of the smallest particles.

The remaining option for drying the sample air is dilution with dry particulate free air. Dilution is also recommended, if the total particle concentration exceeds the single-particle counting range of the CPC ($>30\,000\text{ cm}^{-3}$ in the A20 CPC). The diluter needs to be suitable and characterized for sub-3 nm particles (any turbulent flows in the diluter might lead to high losses). One option designed explicitly for the sampling of nanoparticles is the Airmodus Nanoparticle Diluter (AND), which additionally includes the functionalities of the Kangasluoma et al. (2016a) inlet for core sampling, background measurement and ion precipitation.

In general, when the intended measurement conditions deviate from the limits indicated in the instrument manual (temperature: 15 °C–30 °C, pressure: 90–105 kPa, relative humidity: 0–95%), contacting the manufacturer is advised. This can happen, e.g. during measurements at high-altitude sites or aircraft measurements. In this case, the MFC settings need to be adjusted for the PSM flows to be controlled properly. Assuming an ideal gas, the proper MFC settings (slope) can be calculated using the following equation

$$S_{\text{MFC}} = \frac{T_0 P}{T_{\text{MFC}} P_0} \quad (1)$$

where P is pressure at the measurement site, T_{MFC} estimated temperature of the MFC and T_0 and P_0 are temperature and pressure at which the MFCs were calibrated ($T_0 = 273.15\text{ K}$ and $P_0 = 101325\text{ Pa}$). For temperature, the governing parameter is the temperature of the fluid in the MFC, which is different from the ambient temperature. When shipped from the manufacturer, the slope parameter S is set to correspond to conditions where $T_{\text{MFC}} = 293\text{ K}$ and $P = 101325\text{ Pa}$. Settings need to be changed for both the saturator flow MFC and the excess flow MFC of the PSM using serial commands. In addition, the pressure limits for the self-test (which the instrument performs at startup) need to be adjusted. Otherwise, the instrument will not start. PSM calibrations down to 50 kPa are shown by Kangasluoma et al. (2016a), indicating that variations in the instrument cut-off size due to pressure are negligible, but a pressure-dependency in the dilution correction and counting efficiency of the CPC need to be taken into account when the pressure is lower than ca. 80 kPa.

2.5. Measurement modes

The nCNC software (the current version when writing this is A1X_v2_55) offers three different operation modes: fixed mode, stepping mode and scanning mode.

The simplest operation mode, “fixed mode”, keeps the saturator flow at a constant value selected by the user (between 0.1 and 1.3 lpm). In this operation mode, the cut-off diameter is constant (given that the conditions and particle composition do not change) and the data interpretation of the PSM is similar to any normal CPC measuring the total particle number concentration. This mode also allows the highest time resolution (ca. 1.5 s, [Enroth et al. \(2018\)](#)).

In “stepping mode”, the saturator flow is automatically changed stepwise between two to four different flow rates selected by the user (between 0.1 and 1.3 lpm). The user can select how long the instrument stays at each step. A minimum step time of 30 s is recommended by the manufacturer, as it takes a while (several seconds) for the flow rates and temperatures to stabilize after each change in the saturator flow rate. The stabilization time depends on the difference between the flow rates of successive steps and the response time. Therefore, a part of the data needs to be discarded when operating the PSM in this mode. The Airmodus software discards the first 10 s of data and calculates the median concentration over the rest of the time at each step. The choice of this mode allows for comparing the measured concentration at a few predefined cut-off sizes and generating 1–3 size bins in the sub-3 nm size range.

The third operation mode is the “scanning mode”, in which the saturator flow is continuously and linearly ramped up and down. The user can select the range of flows and scanning time. Typically, we have used a scan time of 240 s for one full cycle from 0.1 lpm up to 1.3 lpm and down to 0.1 lpm. This results in a time resolution of 4 min when the concentration is averaged over the full scan, as the A11 software does. This mode allows the selection of the size bins after the measurements during the data inversion process (see chapter 4).

The consideration on which measurement mode to use can be summarized as following:

- Fixed mode is used for measuring the total particle concentration with a certain cut-off size with a high time resolution. Such cases include total concentration monitoring, operating the PSM as part of a CPC battery or as a detector in an electrical mobility spectrometer ([Kangasluoma et al., 2018](#); [Stolzenburg et al., 2017](#)).
- If the user wants to have size-segregated information, the choice of the mode lies between stepping mode and scanning mode. Scanning mode gives the most information, as in principle all data points can be used, but the data inversion is more complicated. The scanning mode fits best to situations where the process of interest is relatively slow, e.g. studying regional new particle formation, when the aerosol concentration and size distribution do not change considerably over the scan time. In that case, the scanning mode can be used to retrieve the size information of ca. 1–4 nm particles ([Lehtipalo et al., 2014](#)). The exact size range depends on the temperature settings of the PSM.
- If the particle concentrations fluctuate rapidly or the number of sub-3nm particles is very low, it might be more practical to operate the PSM in stepping mode, so that the concentration at each cut-off diameter can be averaged over a longer period of time before comparing the concentrations. Stepping mode data is simpler to post-process, but the time resolution is typically similar (order of minutes) than in the scanning mode.

3. Procedure

3.1. Calibration

The calibration routine for the nCNC consists of concentration calibration for the CPC and cut-off calibration for the full PSM-CPC setup. In the case of long-term operation, at least annual calibration is recommended.

The accuracy of the PSM’s concentration measurement is directly related to the counting accuracy of the CPC, as well as the accuracy of the PSM mass flow controllers as the sample effectively gets diluted due to the mixing of the sample and the saturated flow. The CPC needs to be calibrated for the concentration range that is expected for the total concentration, typically $[10^1 - 10^5]$ ($1/\text{cm}^3$). Note, that in some cases (e.g. during intensive new particle formation or close to traffic), the concentration at the lowest cut-off size might be very high.

The concentration calibration follows standard procedures of a CPC calibration ([Wiedensohler et al., 2018](#)). In short, the concentration calibration is done by comparing the concentration measurement of the test CPC against the concentration measured with a reference CPC or faraday cup electrometer (FCE) for size-selected aerosol particles (e.g. silver).

In the following sub-chapters we will describe the PSM detection efficiency calibration in more detail, as the size range presents additional requirements for the setup compared to typical CPC calibration.

3.1.1. PSM calibration setup

The PSM cut-off size is calibrated using similar overall principles as for any other CPC. An aerosol population with known composition, charge and size is produced and the detection efficiency as a function of particle size is measured against a reference instrument. In addition, if the PSM is used in a scanning mode, the detection efficiency should be measured as a function of the saturator flow rate for each particle size. For stepping mode measurements, the detection efficiency should be measured for each of the saturator flow rate steps to be used.

A differential mobility analyzer (DMA) with high resolving power, $Z/\Delta Z$ more than 20, is needed to achieve a sufficiently monomobile aerosol population downstream of the DMA. For example, the modified Vienna type DMA (so-called Herrmann DMA; [Kangasluoma et al. 2016b](#)) and the commercial Half mini DMA ([SEADM; de la Mora and Kozłowski \(2013\)](#)) are suitable analyzers for the calibration setup. To ensure calibration reproducibility, the DMA sheath flow should be filtered and operated in a closed loop recirculation to avoid water vapor or impurities from entering the aerosol flow. Depending on the particle production method, a neutralizer

might be required to charge the sample upstream of the DMA. An example of a suitable calibration setup is shown in Fig. 4.

There are several means of generating sub-3 nm particles suitable for CPC calibration (e.g. Brilke et al., 2020; Kangasluoma et al., 2013). Ideally, the PSM should be calibrated using the same or similar particle composition that is used later in the real measurements and at the same external conditions. However, usually this is not possible due to the unknown composition of ambient particles and due to technical limitations. Producing a large enough quantity of sub-2 nm particles for the calibration is often the most demanding part of building and operating the calibration setup. In this size range, a FCE is the only instrument, which has a counting efficiency almost independent of the particle size and is thus suitable as a reference instrument. Due to their operating principle, FCEs have higher background noise levels than CPCs and therefore they need higher number concentrations (at least a few hundreds, but preferably a few thousands in $\#/cm^3$) in order to obtain reference concentration with an acceptable signal to noise ratio.

For convenience and repeatability, a glowing wire generator with tungsten or nickel chromium wire is often used to produce charged test particles. This is also the particle generation method used by the nCNC manufacturer. Special attention to the cleanliness of the samples and optimization of the parameters governing the size distribution is required to obtain particles with desired composition and enough signal at the smallest sizes close to 1 nm. In the case of glowing wire generator, this means optimizing the current through the wire and the flow rate of the carrier gas through the generator. When using a tube furnace to produce test particles (e.g. silver or ammonium sulfate), a proper cleaning cycle is needed for producing a clean sample aerosol (Kangasluoma et al., 2013). This requires a two-step cleaning of the tube and vessel: First, using solvents for polar and nonpolar contaminants and afterwards thermal cleaning through pyrolysis in a clean nitrogen atmosphere and temperature above 1000 °C.

3.1.2. PSM calibration procedure and expected results

The actual calibration process starts by tuning the particle generator and scanning the DMA voltage to measure the mobility spectrum of the calibration aerosol, to assess whether there are enough small particles produced. When using a wire generator as the particle source, our empiric experience is that increasing the carrier gas flow rate brings the particle mode towards smaller particles and increasing the wire temperature increases the signal magnitude.

The next step is to select a fixed voltage for the DMA corresponding to the desired size. We normally select a small size (e.g. 1.2 nm in mobility diameter) first to check if the instrument is able to activate the smallest particles. If needed, the operating temperatures of the PSM can be adjusted at this point. Increasing the difference between the saturator and growth tube temperature increases the detection efficiency for small particles. However, the background level (counts resulting from homogenous nucleation) should be measured after changing the settings so that it does not become too high (see chapter 3.2.3). Optimal settings are specific to the instrument and the desired measurement range.

Next, the concentration is measured for a range of fixed DMA voltages. About 10–15 sizes in the size range of about 1–4 nm (depending on the PSM settings) are needed for characterizing the detection efficiency and activation curves. For a scanning mode calibration, the PSM is set to scan at least 4 full saturator flow scans at each voltage. The selected particle diameter is increased until the PSM's detection efficiency is not any more dependent on the saturator flow rate, i.e. particles are activated already with the smallest saturator flow. During the calibration, the wire current and flow rate typically need to be adjusted a few times to ensure that the number concentration does not exceed 10 000 ($1/cm^3$). This way, the concentration stays well within the CPCs single particle counting range.

The detection efficiency is calculated by dividing the concentration measured by the PSM with the one measured by the electrometer. For particles in the sub-5 nm size range, the multiply charged fraction is negligible (Wiedensohler & Fissan, 1991). However, the possible background for each instrument needs to be measured and accounted for, in case the number concentration of calibration particles is low. For fixed mode calibration, the results are plotted against particle size and a typical cut-off curve is achieved (Fig 5). From stepping/scanning mode calibration, a similar plot can be made for each saturator flow rate. It is typical for the PSM/nCNC that the detection efficiency does not reach 100% even at sizes larger than 3 nm, but it reaches a “plateau value” in the range of 70–90%. The main reason for this saturation behavior is turbulent losses of particles in the PSM mixing section (Vanhanen et al., 2009).

For the analysis of scanning mode data, we are also interested in the saturator flow rate at which different sized particles activate, so called “activation saturator flow rate”. To determine this, we can find the saturator flow rate, where the detection efficiency reaches half of its maximum value for each particle size (Fig. 6A). The maximum and half-maximum points can be derived from the calibration data via interpolation or with a suitable fit function, which has these values as fitting parameters. Prior to analyzing the scan mode data, the time lag between the moment when the mass flow controller settings are changed (which changes the saturator flow rate) to the moment when a particle reaches to the detector needs to be corrected. The time lag is about 3 s for the standard PSM setup.

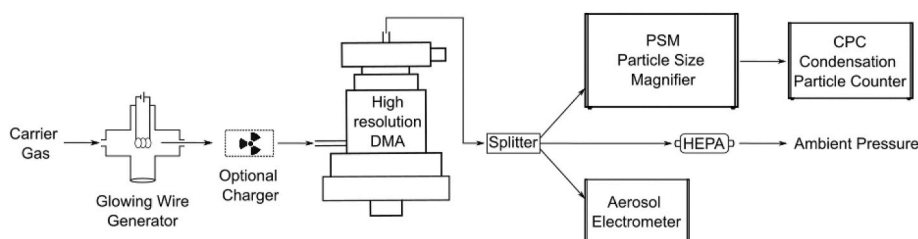


Fig. 4. An example of a possible calibration setup for the PSM/nCNC.

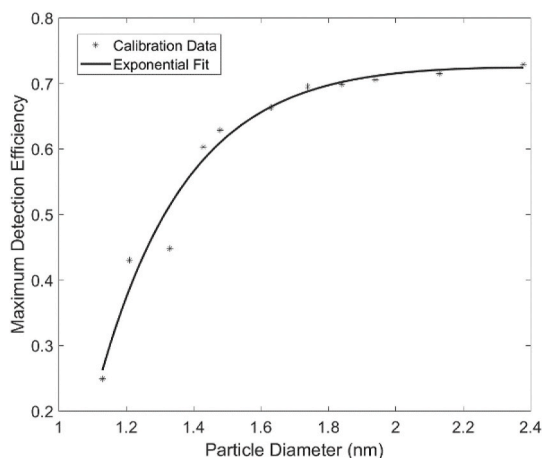


Fig. 5. An example of the PSM's detection efficiency as a function of particle diameter at saturator flow rate 1.3 lpm.

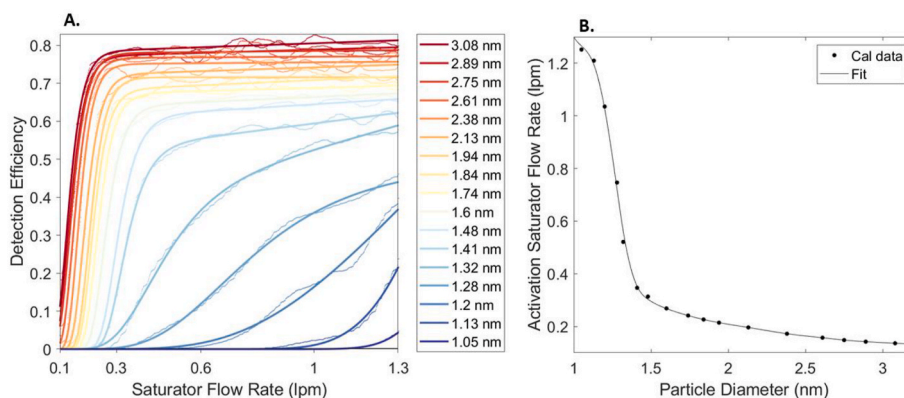


Fig. 6. An example of the PSM calibration results in the scanning mode. A) gives detection efficiency as a function of saturator flow rate for different particle sizes. The thin line gives the actual data points and the thicker lines are fits to the data points. B) gives the activation saturator flow rate as a function of particle size.

The activation saturator flow rates derived from Fig. 6A are plotted as a function of particle diameter in Fig 6B. It can be seen that the activation size is not linear as a function of the saturator flow rate. When the saturator flow rate is increased from its smallest values, the activation diameter decreases very fast in the beginning, but after ca. 1.5 nm, the decrease slows down. Due to this behavior, it is advised to have more calibration points in the steep part of the calibration curve. The saturator flow-activation size curve is useful also for selecting suitable saturator flow steps for the step mode operation of the PSM.

3.1.3. Uncertainties in the PSM calibration

There are several unsolved issues in the calibration of sub-3 nm particle counters that add to the uncertainty of the measurements. The first problem is that the only available reference instrument (FCE) and particle size selection method (DMA) rely on the electric charge of the particles. Thus, we can only calibrate the PSM using charged particles (typically negative particles are used), although in the field it is used to measure both neutral and charged particles. There are studies indicating that the activation diameter of neutral and charged clusters of similar composition can vary by up to 0.5 nm (Kangasluoma et al., 2016c). This is because charge lowers the energy barrier for heterogeneous nucleation and thus charged particles activate at lower supersaturations than neutral particles (e.g. Winkler et al., 2008). New computational studies show that the relevant factor governing the activation probability is the amount of hydrogen bonding between the seed particle and the vapor, which is affected by the particle charging state, as well as the composition (Keshavarz et al., 2020a,b).

This brings us to the second problem, which is that the particle composition affects their activation probability. In some cases, it is possible to tailor the calibration aerosol to have a similar composition as is expected from the real measurements (Ahonen et al., 2017), which minimizes the composition uncertainty. Another option is to compare the PSM measurement to an instrument with a different operating principle and determine the shift in the cut-off size for a certain type of particles compared to the calibration particles. This method has been used e.g. in chamber experiments, where the particle composition was known, but a suitable calibration aerosol was not available beforehand (Dada et al. (2020); Rörup et al. (2021)). However, the ambient particle population might be a varying

mixture of different compositions, so the composition effect is difficult to solve and characterize.

The third problem is related to the particle carrier gas. In calibrations, dry nitrogen or filtered air is usually used as carrier gas of the particles. However, in field conditions, the sample RH is typically higher, which may result in higher background level and higher detection efficiency (Rörup et al., 2021). One way to overcome this is to monitor the background level during calibration and adjust the saturator temperature in field conditions to match the background during calibration in order to achieve a detection efficiency close to that of the calibration. Jiang et al. (2011) show that the amount of background counts is a good indicator of the supersaturation level in the instrument. A preferable option would be to perform the calibration at RH close to the measurement conditions and control the sample humidity to the calibration RH, but this is often technically difficult.

In summary, as the PSM measurement principle is based on particle activation, one should always keep in mind that the reported diameters are “activation equivalent diameters”, analogous to e.g. mobility equivalent diameters. This means that we assume that the measured particles activate in the PSM in a similar way than the calibration aerosol. This can lead to significant uncertainty for different aerosol types and when comparing to instruments with different operation principle. Thus we recommend, that the material and polarity of the particles used for calibration, as well as the RH of the calibration aerosol is always reported together with the data and results from the PSM/nCNC.

3.2. Maintenance and verification during normal operation

Here we describe the maintenance tasks that should be performed when using the PSM/nCNC in long-term field measurements. Table 1 summarizes the frequency of the maintenance routines. Some of them can be automated with a suitable setup. Additionally, we recommend monitoring the status of the instrument (possible warnings or error messages) and plotting the data daily. Supplementary materials to this article presents a troubleshooting table, which can be of help when encountering problems with the PSM/nCNC.

3.2.1. Filling and draining

The PSM's fill bottle should always be connected when performing measurements. The flow of DEG into the saturator is controlled by the hydrostatic pressure from the liquid in the fill bottle. Therefore, it is necessary that the bottle is above the PSM, but at the same time not too far above (see section 2.3.1). Be careful not to mix the PSM and CPC fill bottles (use properly labeled bottles), as any DEG going into the CPC will contaminate it.

After installation, the empty fill line needs to be “primed” prior to use, to allow the liquid to reach the saturator. This is because DEG is quite viscous and does not flow into the saturator when there are large air pockets in the line. Priming is done using the PRIME function from the software, which opens a valve behind the saturator allowing liquid to move freely in the line. The default priming time is 90 s. The operator should ensure that the liquid is flowing in the line while priming. This can be seen by monitoring the saturator temperature, which drops for a while, when new DEG is flowing into the saturator.

During normal measurements, DEG flows slowly through the saturator keeping the porous medium in the saturator wetted. Excess DEG is collected into a drain reservoir where two liquid level sensors prevent overflowing the reservoir. This drain reservoir needs to be emptied, “drained”, typically about once a week (Table 1) but latest when the instrument reports a warning due to high liquid level. The need for draining is more frequent in environments where gas-phase water concentration is high, i.e. warm humid environments.

When the liquid level reaches the lower of the two sensors, the instrument gives a warning for high liquid level with yellow indicator (HIGH) in the front panel, as well as in the software. The warning does not affect the measurement or data quality. When the liquid level reaches the second sensor, the instrument shows a red indicator (FULL), and it goes into standby mode to prevent overflowing the PSM and the CPC with the working fluid. In standby mode, the MFCs and the temperature control are turned off and the valve in the fill line is closed. To recover from the standby mode, the PSM needs to be hard-booted from the power switch after the instrument has been drained.

Before the instrument is shipped or in case it needs to be tilted or moved, it needs to be dried thoroughly to avoid having sensitive parts wet. Drying starts by draining the instrument properly and then letting it run in DRY mode for a minimum of 2–4 h. More instructions on draining and drying are given in the instrument manual.

As the PSM's sample air is typically not dried, some water may also accumulate in the CPC. To avoid this, it is advised to also drain the CPC regularly (e.g. weekly). The required frequency depends on the temperature and humidity of the sampled air in the measurement location.

Table 1
Summary of the regular maintenance tasks for the PSM/nCNC.

Maintenance tasks	~Daily	~Weekly	~Yearly
Adding DEG and butanol to fill bottles		x	
Draining the PSM and CPC		x	
Cleaning drip bottle and lines		x	
Manual inlet flow measurements		x	
Background measurements	x ^a	x	
Calibration			x

^a If operated at background $>0 \text{ cm}^{-3}$.

3.2.2. Flow verification

The sample flow rates of the PSM and the CPC need to be verified upon installation. During long-term measurements, these should be monitored regularly. We recommend at least weekly manual flow rate check. If the flow rates clearly deviate from their nominal values, it is typically an indicator of an issue that needs to be fixed (see Troubleshooting in the Supplementary materials).

The CPC sample flow rate directly affects the number of particles counted by the instrument, and thereby the concentration reading. A difference between the actual flow rate and the flow rate specified in nCNC control software will cause a systematic error in the particle number concentration. The volumetric flow rate of the CPC can be measured by connecting a low-pressure-drop flow meter (e.g. DryCal or Gillibrator) to the CPC inlet. The flow rate can also be measured with mass flow meters, e.g. hot wire sensor-based meters and making the conversion into volumetric units. When using mass flow meters, it is important that the meter has settled into the same temperature as the inlet flow rate.

The nominal inlet flow rate for the A20 is 1 lpm; values between 0.98 and 1.09 lpm are considered normal. The measured flow rate should be updated manually to the instrument software. The flow rate through the CPC's critical orifice often decreases with time. If the flow rate decreases significantly, it may imply that the critical orifice needs cleaning. This can also be indicated by the critical orifice warning on the panel screen of the CPC and software.

The nominal inlet flow rate of PSM is 2.5 lpm and the normal values are in the range 2.5–2.6 lpm. A deviation from this range might be an indicator of several issues, for example leaks in the internal lines or in the line between PSM and CPC. Note, that the CPC needs to be connected to the PSM when measuring the inlet flow rate. If the CPC's sample flow rate is incorrect in the nCNC control software, it can also cause deviation in the PSM inlet flow rate.

3.2.3. Background measurements

To increase the activation efficiency of smallest particles, the PSM is sometimes operated at settings where homogenous nucleation of DEG is allowed at the highest saturator flow rates (this is called “background”). The tolerated background level depends on the application and environment (expected particle concentration in the sample and the supersaturation needed for activating the particles of interest). For example, when measuring in a boreal forest, we found that if the background is very low ($<1 \text{ cm}^{-3}$ at 1.3 lpm saturator flow), we were not able to detect most of the presumably organic clusters smaller than 2 nm present in the forest (Sulo et al., 2021). For organic particles, background levels as high as $1\text{e}1$ to $1\text{e}2 \text{ cm}^{-3}$ might be required to activate sub-2nm particles, while smaller supersaturations are required for inorganic particles (Rörup et al., 2021). However, in other applications, like using the PSM as a detector in a mobility spectrometer, it might be important to keep the background at 0 cm^{-3} .

Regular monitoring of background and its changes also gives an indication of the activation efficiency of the instrument. Even if the instrument is used at zero background level, we recommend measuring the background e.g. weekly in connection with the flow verification, as an increasing background level could indicate problems or that the instrument lines need to be cleaned.

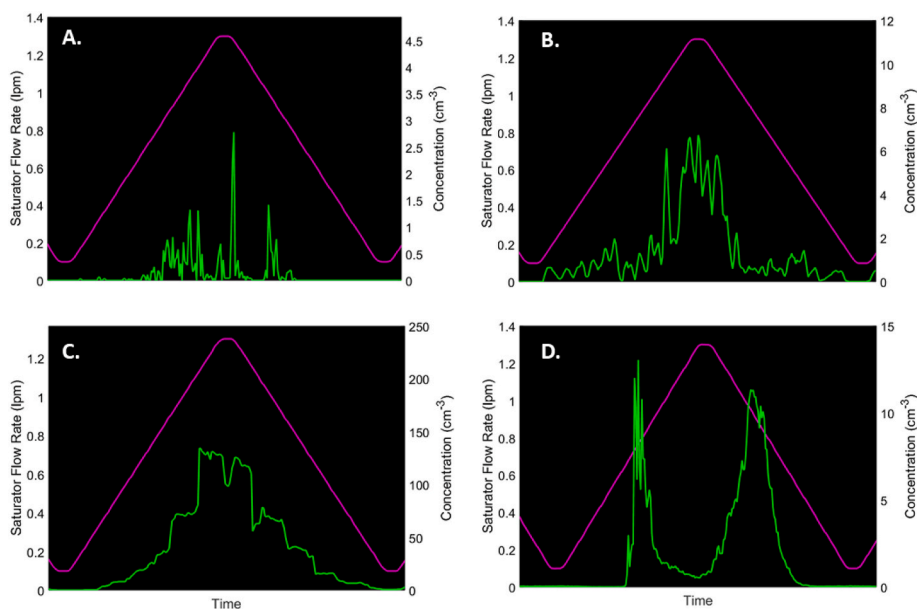


Fig. 7. Examples of PSM background measurements in scanning mode at different settings. The pink line shows the saturator flow rate (left axis) and the green line is the measured concentration (right axis) during a single scan. Panels A–C shows examples where the background is close to zero at low saturator flow rates and is increasing with increasing saturator flow rate, as it should. The supersaturation and thus amount of background counts is increasing from A to B to C. Panel D shows an example with “side peaks” in the background measurements, indicating homogenous nucleation in the mixing section, which should be avoided when using the scanning mode. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

In case the PSM is used with high sensitivity settings (allowing a certain amount of background counts), the background should be monitored with proper machine-readable time stamps so that the counts from homogenous nucleation can be subtracted during data analysis. This can be automatized and incorporated into the inlet control system using a setup presented in Fig 3. We recommend at least three to four 12 min intervals of background measurement per day initiated at random times. This will provide the background data that is randomly distributed to the operation hours allowing to determine non-biased diurnal number concentration measurements at the observation site.

Background measurement is a concentration measurement with particle free air (zero-air) closely representing the ambient gas phase composition. In practice this means filtered outdoor air when measuring atmospheric aerosols. Having a continuous flow of ambient air through a filter, keeps the filter in an equilibrium with outdoor air, for example with respect to gas phase water concentration. This is not entirely true for sticky gases, but it is still a reasonable representation of ambient air. Manual background measurements can be done by simply placing a HEPA-filter in front of the PSM inlet. Do not disconnect the PSM from the outside inlet and place the filter inside the measurement cabin, as the indoor air might have different composition and temperature as the outdoor air, affecting the background measurement. Instead, if possible, put the filter in front of the outside inlet.

There are several things to check from the background measurements:

- The background should be approximately constant in time. Increasing background can indicate problems in the instrument (especially if the background increases suddenly). A decreasing background could indicate that the instrument is no longer able to activate the smallest particles. Note, that changes in the gas phase composition (especially water content) can also affect the background.
- The concentration should be zero at the smallest flow rate (close to 0.1 lpm), corresponding to the largest cut-off size. If this is not the case, there might be liquid DEG in the lines between the growth tube and CPC. This can be fixed by flushing the line from PSM's inlet to outlet with clean air (see next chapter). Another reason for the high background at the smallest flow rate might be a leak inside the PSM, which means that some of the sample air is taken from along the internal lines and not from the inlet.
- The number of counts from homogenous nucleation should increase as a function of saturator flow rate and the maximum number of counts should occur with the highest saturator flow rate (Fig. 7). Sometimes homogenous nucleation of DEG might happen in the mixing section of the PSM (Kangasluoma et al., 2016a) causing additional maxima on the rising and falling part of the scan (between 0.3 and 0.8 lpm saturator flow rate, see Fig. 7D). This makes the analysis of scanning mode data impossible. The intensity of these unwanted peaks can be reduced by decreasing the temperature difference between the inlet and the saturator. If this does not help, the mixing section and/or the saturator need to be cleaned (usually by manufacturer).

3.2.4. Cleaning excess DEG from the lines

Sometimes liquid may accumulate into the line between the PSM and the CPC due to condensation of water and DEG. This liquid causes background counts whose number is independent of the saturator flow rate. Furthermore, if the liquid reaches the CPC optics, it can lead to premature fouling of the optics and the decrease of the CPCs performance. A drip bottle that acts as a liquid trap at the outlet of PSM decreases the probability of liquid reaching the CPC (Fig. 2B).

The DEG trap needs to be emptied from time to time, but well before it is full. Even with the DEG trap, there can be some excess liquid accumulating in the lines. Therefore, we recommend cleaning the tubing between PSM and CPC regularly (e.g. weekly) to prevent problems. The line can be cleaned by blowing air through it or rinsing it with water (when disconnected from the instrument).

If cleaning the PSM-CPC line does not solve the background problems, the PSM can also be "flushed" with air to remove extra liquid in the internal lines. Flushing can be performed by first disconnecting the inlet and outlet of the PSM and pointing a compressed air line partially towards the inlet. At the same time, e.g. a paper towel is brought to the outlet to catch the liquid coming out. The flow rate does not need to be extensive, and in no case should the compressed air line be pressed fully against the inlet tube. The purpose is to flow air through the instrument but not to increase the pressure inside it. The flow through the instrument should be continued until there is no more liquid coming from the outlet.

Table 2

The most important diagnostic parameters to monitor.

Diagnostic parameter	Explanation	Normal range
PSM status	1 = ok, 0 = not ok	1
PSM note	1 = ok, 0 = warning	1
CPC liquid level	1 = ok, 0 = not ok	1
Δp_{crit}	Pressure drop over the critical orifice of the CPC	55 kPa ^a >
Δt_{pulse}	Average pulse duration in CPC (only at concentrations below 5000/cc)	0.3–0.6 μ s
$\Delta p_{psm} \approx p_{abs,psm} - p_{abs,cpc}$	Pressure drop in PSM	1–2 kPa
Δp_{nozzle}	Pressure drop in CPC's nozzle	1–5 kPa
T_{gr}	PSM growth tube temperature	setpoint value
T_{sat}	PSM saturator tube temperature	setpoint value
T_{drain}	PSM drainage temperature	setpoint value
$Q_{sat,psm}$	PSM saturator flow rate	setpoint value

^a Depends on the ambient pressure. The minimum is ~55% of ambient pressure.

3.2.5. Data monitoring

We recommend daily monitoring the nCNC operation to detect any problems and ensure data quality. Near-real-time data monitoring should include graphical presentation of raw concentrations, inverted concentrations, background measurement and the time series of diagnostic data such as instrument flow rates, pressures and temperatures (see Table 2). If a value is out of its normal range, we suggest consulting the nCNC manual and/or the troubleshooting table in the Supplement to solve the issue.

4. Data treatment

4.1. Data pre-processing

The raw data from the nCNC is recorded at 1-s intervals. The concentration given by the A11 program is the measured total particle number concentration ($\#/cm^3$) at the current saturator flow rate, corrected for the dilution inside the PSM.

The concentration measured by the CPC needs to be corrected for dilution. Dilution occurs when the sample flow is mixed with the particle-free saturator air inside the PSM, thus the actual concentration can be calculated with the following equations:

$$N_{nCNC} = N_{CPC} * \text{Dilution ratio} \quad (2)$$

$$\text{Dilution ratio} = \frac{Q_{inlet} + Q_{saturator}}{Q_{inlet}} \quad (3)$$

where N_{nCNC} is the corrected concentration, N_{CPC} is the concentration recorded by the CPC and Q_{inlet} and $Q_{saturator}$ are the flow rates through the inlet and saturator of the PSM, respectively. N_{CPC} , the dilution ratio and the flow rates are also recorded in the raw data file, should there be a need to recalculate the concentration.

For the removal of background counts (in case the instrument is operated at background $>0 \text{ cm}^{-3}$), the measured background is averaged over each background measurement period for each saturator flow rate and then interpolated between successive background measurement periods. These counts are then subtracted from the measured concentration before further analysis.

In case the PSM is used in fixed mode, the nCNC data analysis follows otherwise exactly the same principles as any other CPC data. However, one should remember that the maximum detection efficiency of the PSM is typically less than 100%, so the data should be corrected with the plateau value determined from the calibration (Fig. 5). The step mode or scan mode data is typically inverted into a particle size distribution (see chapter 4.3).

When the PSM is used for size distribution measurement, the diffusion losses for particles in each size bin can be calculated using the average diameter of the size bins. Particle losses due to diffusion increase rapidly as the particle diameter becomes smaller. Therefore, it is important to correct for the diffusion losses during data processing, as it can make a substantial difference for the smallest size classes in case the sampling is not ideal. Depending on the Reynolds number for the flow in the sampling line, one can estimate the losses using the Gormley and Kennedy laminar model (Gormley & Kennedy, 1948). In case of a complicated inlet design, it is better to experimentally characterize the losses as a function of particle size in the sampling lines and use these values to correct the final concentrations. A code to estimate sampling losses in a core sampling system is given by Fu et al. (2019).

4.2. Quality assurance

During data quality assurance, the values associated with an error code from the nCNC software should be removed, as they point towards a technical problem affecting the data quality. The values with a warning should be flagged with the warning code and the

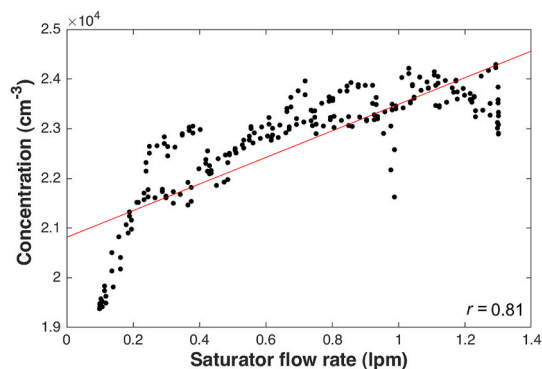


Fig. 8. An example of the measured number concentration plotted against the saturator flow rate during one full scan (from 0.1 to 1.3 lpm and back to 0.1 lpm). When particles smaller than 3 nm are present, the number concentration increases with saturator flow rate (decreasing cut-off size). The remaining variation is caused by random fluctuations in the concentration. The red line indicates the line of best fit. The Spearman's rank correlation coefficient is 0.81, so the scan passes the quality control algorithm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

data points should be used with caution.

The background level can be used as an additional quality assurance parameter. If the background is very high compared to the expected signal or it has suddenly clearly increased compared to normal level, the data should be inspected closely or discarded. When the PSM is used in scanning mode, the background should always be zero at low saturator flow values (0.1–0.2 lpm) and increase as a function of the saturator flow rate (as supersaturation increases). Other kind of behavior points towards a malfunctioning of the instrument (see chapter 3.2.3).

As the size distribution from the PSM is determined from the difference in total concentration measurement between consecutive measurement points at different saturator flow rates, any changes in the total concentration during the measurement will interfere with the method. To remove scans with significant changes in the total concentration, Chan et al. (2020) developed a quality assurance algorithm checking the shape of the scans. The algorithm checks that the measured concentration in each scan has a positive and statistically significant correlation with the saturator flow rate (see Fig. 8). This means that the concentration increases with decreasing cut-off diameter, as it should when 1–3 nm particles are present in the sample. Scans that do not fulfill these criteria (concentration decreases or does not correlate with saturator flow rate) are replaced with NaN values, as they would lead to unphysical results and noisy data. Typical reasons for discarded scans are changes in air masses occurring during the scan due to wind shifts or local pollution spikes that are faster than the scan time. Even though a scan is discarded from size distribution analysis, it can be used for determining the total concentration above certain cut-off size (e.g. > 4 nm at saturator flow rate of 0.1 lpm).

The above-mentioned method should work for background stations and other measurement locations, where changes are typically slow and a population of sub-3nm particles is constantly present. For measurement locations with significant local sources (e.g. traffic), the method may discard a large fractions of scans due to rapid changes in the signal. One might try averaging over several scans or smoothing the data to remove some of the variability before further processing the scanning mode data. Another solution is to use the PSM in step or fixed mode instead of scan mode or using another CPC to record the variability in the concentration of the larger particles during the scanning time. For further discussion about the PSM data preprocessing and quality assurance we refer the reader to Chan et al. (2020).

4.3. Data inversion and typical results

A data inversion is required to retrieve the particle size distribution from the PSM data. The inversion is based on the relationship between the saturator flow rate, particle diameter and detection efficiency, which is established during the calibration (see chapter 3.1.2). Using this information, the measured particle concentration as a function of saturator flow rate can be converted into a size distribution, assuming that the measured particles activate inside the PSM in a similar manner as the calibration aerosol. Therefore, the size given by the PSM is always activation-equivalent-diameter. Due to uncertainties in the calibration (see 3.1.3), the sizing might not be as accurate as for best electrical mobility spectrometers (EMs), but, on the other hand, the PSM can provide size information at smaller sizes and lower concentrations than EMs, which suffer from low charging probability and high particle losses, especially at sub-3 nm sizes (Kangasluoma et al., 2020).

At least four different inversion methods have been applied to PSM data measured in scanning mode: step-wise inversion and kernel function inversion were introduced by Lehtipalo et al. (2014) and used in many following studies. Later, Cai et al. (2018) proposed using Hagen and Alofs method (H&A (Hagen & Alofs, 1983); or the expectation and maximization algorithm (EM (Maher & Laird, 1985);). Chan et al. (2020) compared all of those four methods using test data from an urban site in Beijing.

The step-wise method is the inversion method used by Airmodus Ltd and included in the A11 software. This method assumes that the size resolution of the PSM is infinite (i.e. that the cut-off size is always a step function); and as such, there is a one-to-one relationship between the particle diameter and the saturator flow rate. Hence, the total number concentration in a specific size range can be determined by differentiating the measured number concentration between the saturator flow rates corresponding to the upper and lower edges of the size bin. The concentration is further corrected by the measured detection efficiency at the middle point of the size bin. The step-wise inversion method is easy to understand and apply, and it provides good results if the cut-off curves are sharp and when not too many size bins are used. Similar method is often used when measuring with a CPC-battery (several CPCs at different cut-off sizes measuring in parallel (Kulmala et al., 2007)), or when the PSM is used in step mode.

The kernel function method and H&A method use measurement-based kernel functions of the PSM. The kernel functions are obtained based on the instrument calibration with monodisperse aerosol and they take into account the shape of the cut-off curves. The number concentrations in each size bin are determined by solving a non-homogenous equation (e.g. with least-squares method), which relates the saturator flow rate to the number concentration. The main difference between the kernel function and H&A method is the number of assumed particle size bins. The kernel function method uses a size bin number much less than the number of saturator flow rates, whereas the H&A method uses a size bin number higher than the number of saturator flow rates. The latter method then reduces the size bin number equal to the number of saturator flow rates via interpolation. The EM method algorithm is an iterative algorithm based on the theories of probability that obtain the maximum-likelihood estimates of particles to be in a particular size bin when the size distribution is not known.

As the inversion methods are still under development and testing, we do not take a stand on what is currently the best method but instruct the user to test the suitability. Chan et al. (2020) found that all of the methods performed rather well during new particle formation events (i.e. when the concentration of sub-3nm particles is expected to be high), but the H&A and EM methods were more robust when inverting noisy data. For further information on these inversion methods, see the studies of Cai et al. (2018); Hagen and Alofs (1983); Lehtipalo et al. (2014); Maher and Laird (1985) and Chan et al. (2020). The current versions of the inversion codes are openly available at https://github.com/Airmodus/A11_Scan and <https://github.com/tommychan-dev/PSM-Inversion>.

An example of final processed data from a PSM measuring ambient air is presented in Fig. 9. Note, that here we have chosen to present the data between 1.3 and 2.7 nm with three size bins, which is a reasonable size resolution given the uncertainty in the absolute diameters during ambient measurements. The data could be inverted also to a finer size resolution, which might be useful e.g. for laboratory experiments, as the uncertainty in particle diameter is smaller when the particle composition and charge is known. More discussion on the sources of uncertainty in sub-10nm size distribution measurement, including supersaturation scanning methods, can be found in the article by Kangasluoma et al. (2020).

4.4. Quality control

After data processing and inversion, the data should still be quality controlled. Concentrations should remain positive and any values that differ substantially (e.g. more than 3 standard deviations) from the median concentration should be verified before use. A reasonable upper limit value for the concentrations depends on the environment and proximity to sources of particles.

The data should have a reasonable daily and seasonal cycle, depending on location. Higher concentrations of sub-3 nm particles are often observed during daylight hours and spring/summertime in forest environment and during rush hours in cities (Kontkanen et al., 2017; Zhou et al., 2020). In some environments, even a majority of total particle concentration can be in sub-3nm size range (cluster mode) (Kontkanen et al., 2017; Zhou et al., 2020), while some environments show very low number of clusters (Jokinen et al., 2018; Yu et al., 2013).

In practice, it is also useful to compare the total number concentration measured by the PSM to another particle counting instrument (e.g. number concentration from another CPC or integrated concentration from an electrical mobility spectrometer, EMS), if available. The size distribution from the PSM in scanning mode can be compared to ion size distribution (e.g. from air ion spectrometer) or combined with a size distribution from an EMS or neutral cluster and air ion spectrometer (NAIS (Mirme & Mirme, 2013)). However, it should be noted that the overlapping size range (ca. 2.5–4 nm) is often problematic for all of the above mentioned instruments, as the PSM sizing gets less accurate above 3 nm (Cai et al., 2018), while NAIS particle mode suffers from background due to corona charger ions at sizes < 3 nm (Manninen et al., 2011) and electric mobility spectrometers have very poor detection efficiency (Jiang et al., 2011b). Therefore, a perfect match between the number concentrations measured with each instrument is currently difficult to obtain, especially in field conditions. Kangasluoma et al. (2020) discussed and compared the different methods currently available to measure sub-10 nm size distributions and evaluated their pros and cons.

5. Conclusions

Measurement of nanoscale aerosol particles is challenging especially in the sub-3 nm size range due to their low charging probability and high diffusional losses, which limit the applicability of electrical measurement techniques. Condensation particle counter-based methods, like the PSM/nCNC, provide an alternative method to detect and size analyze atmospheric clusters and particles. The PSM/nCNC has already produced invaluable new data about particle formation and early particle dynamics in different kinds of environments and chemical systems leading to significant scientific breakthroughs, as well as shown that sub-3nm clusters are formed or emitted from a range of different kinds of processes and equipment and are thus almost omnipresent in our atmosphere.

This procedure is a step towards harmonizing and standardizing field measurements of sub-3 nm particles using the PSM/nCNC. The inclusion of sub-10 nm aerosol measurements in international observation networks, such as the ACTRIS station network, puts high demands on data quality and comparability. The guidelines and recommendations presented here can, for most parts, also be utilized in other applications than atmospheric observations, such as indoor air measurements, chamber experiments or fundamental cluster research. By improving the accuracy and availability of nanoparticle measurements, we can improve our understanding of the role of aerosol formation on climate and air quality and increase the fundamental knowledge of cluster and nanoparticle processes.

There are still challenges in the measurement of sub-3nm aerosol, which complicate the interpretation of PSM/nCNC data: a) the particle composition and charging state varies and the sample is often a mixture of different particle types, affecting their activation probability, b) the PSM is usually operated at high supersaturations at the verge or slightly over the onset of homogenous nucleation, which is a process highly sensitive to temperature and RH, making the instrument sensitive to external conditions, c) the clusters and nanoparticles may undergo rapid physical or chemical processes either in ambient air or during sampling (e.g. evaporation, condensation, clustering) that might affect their concentration, composition or structure, d) sampling sub-3 nm particles with high enough efficiency is often a technical challenge.

While a lot of work has been carried out to characterize these effects, and some of them can be overcome by following standard measurement procedures like the ones presented here, some are related to the intrinsic properties of clusters, so they cannot be totally avoided in any measurement system for nanoscale particles. The PSM/nCNC is currently the most sensitive and best characterized instrument for measuring sub-3nm aerosol particles. New technical solutions and better understanding of cluster processes will further improve measurement practices and data accuracy in future.

Data availability

Matlab codes for visualizing raw data and diagnostics data are available at: https://version.helsinki.fi/baalbaki/psm_diagnostics_plot.

Codes for inverting raw data are available at: https://github.com/Airmodus/A11_Scan (Scilab) and <https://github.com/tommychan-dev/PSM-Inversion> (Matlab).

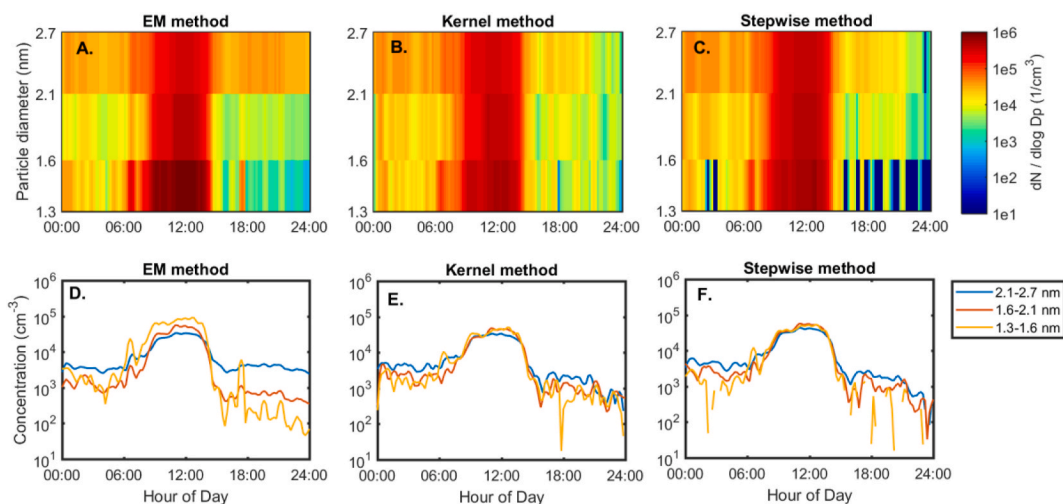


Fig. 9. An example of PSM data from a measurement site in Beijing on 31 Jan 2018 using three different data inversion methods: expectation-maximization algorithm (A, D), the kernel method (B, E) and the step-wise method (C, F). This particular day exhibited strong new particle formation with peak concentration around midday.

Declaration of competing interest

EM and JV are employed by Airmodus. JV, MK and KL are shareholders at Airmodus. The rest of the authors declare that they have no competing interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jaerosci.2021.105896>.

Author contributions

KL and TJ coordinated the work. All authors contributed to the writing and editing of the manuscript. All authors have approved the final article.

About this article

This article is an Editor-Invited Tutorial Article. Tutorial articles, established to commemorate the 50th Anniversary of the Journal of Aerosol Science in 2020, are intended to serve as educational resources for the aerosol research community on state-of-the-art experimental, theoretical, and numerical techniques in aerosol science.

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Jonathan Duplissy is senior scientist at INAR and Helsinki Institute of Physics in Finland. He previously worked at CNRS in France, CSIRO in Australia, PSI in Switzerland and CERN in Switzerland. He completed his PhD in 2008 at ETH Zürich in Switzerland, and he received in 2019 a title of Docent in atmospheric science from University of Helsinki, Finland. He has participated and organized various field and flight campaigns, used simulation chambers (Smog chamber at PSI and CLOUD chamber at CERN), and developed aerosol instrumentations such as HTDMA, AND, PINci, and the CLOUD chamber. His current main research interest is in ice nuclei properties.



Elina Miettinen is application engineer at Airmodus. She completed her MSc (Tech) at Tampere University of Technology in 2012, where she worked in the aerosol physics laboratory, studying production of test aerosols using a portable flame generator. At Airmodus she performs instrument calibrations and training, process development and quality assurance, and supports customers in the installation and operation of PSM and particle counting systems in different applications according to their research and measurement needs.



Joonas Vanhanen is the CTO and co-founder of Airmodus Ltd., a company manufacturing instruments for detecting nano-sized particles in the air. He received his PhD degree from the University of Helsinki in 2018 focusing on new instrument development for detection of freshly formed aerosol particles in the atmosphere. Before Airmodus, he worked as a research scientist at the University of Helsinki as well as at the Finnish Meteorological Institute. His research focused on experimental aerosol science in lab and in field conditions



Juha Kangasluoma is senior scientist at INAR and leads the Aerosol Science and Technology group. He is also a scientist at Karsa Ltd. He completed his PhD at University of Helsinki in 2015 and was a visiting scientist at Beijing University of Chemical Technology in 2018–2019



Markku Kulmala is director of INAR. He completed his PhD at University of Helsinki in 1988 and has been professor since 1996. He has been visiting professor at University of Vienna and University of Stockholm and has several honorary professorships. He is ERC AdG holder, academician and recipient of numerous awards.



Tuukka Petäjä is professor in experimental atmospheric sciences at INAR. He completed his PhD at University of Helsinki in 2006 and was a post-doctoral researcher at NCAR in Boulder in 2007–2008. He is a recipient of Vaisala award for instrument development and Science and Technology in Society Future Leader by New York Academy of Sciences. He is Highly Cited Scientist since 2014.



Tuija Jokinen is assistant professor at the Cyprus Institute Climate & Atmosphere Research Center in Nicosia, Cyprus. Formerly she acted a head of unit for Aerosols, Trace gases and Clouds Research Infrastructure (ACTRIS) calibration centers for in-situ measurements of sub-10 nm aerosol particles (Cluster Calibration Center, CCC) and aerosol precursor measurements (CiGAS-UHEL) and a group leader for the Polar and Arctic Atmospheric research group. She has a MSc degree in analytical chemistry (2008) and a PhD in atmospheric sciences (2015) completed at the University of Helsinki and she has worked at TROPOS in Leipzig, Germany as a visiting researcher and at the University of California as a postdoc. She is recipient of the Finnish Academy postdoctoral scholarship and a University of Helsinki dissertation award (2015).