



# Simulation-aided characterization of a versatile water condensation particle counter for atmospheric airborne research

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## 10 Abstract

Capturing the vertical profiles and horizontal variations of atmospheric aerosols often requires accurate airborne measurements. With the advantage of avoiding health and safety concerns related to the use of butanol or other chemicals, a water-based condensation particle counter (wCPC) has emerged to provide measurements under various environments. However, the airborne deployment of wCPC is relatively rare due to the lack of characterization of wCPC performance. This study investigates the performance of a commercial "versatile" water CPC (vWCPC Model 3789, TSI) under low-pressure conditions. The effect of conditioner temperature on wCPC performance at low pressure is examined through numerical simulation and laboratory experiments. We show that the default instrument temperature setting of 30°C for the conditioner is not suitable for airborne measurement and that the optimal conditioner temperature for low-pressure operation is 27 °C. Additionally, we show that insufficient droplet growth becomes more significant under the low-pressure operation. The variation in the chemical composition can contribute up to 20% uncertainty in the counting efficiency of the wCPC, but this variation is independent of pressure.

## 1 Introduction

Atmospheric aerosol particles are one of the key components of the atmosphere. The currently known ambient aerosol has a size range over several magnitudes and consists of complex chemical compositions, which vary with size, origin, age, and atmospheric processing. This tiny but complicated particulate matter plays a remarkable role in climate change (Seinfeld et al., 2016) and human health (Anderson et al., 2020; Lighty et al., 2000; Pöschl, 2005). To understand the variation of atmospheric aerosol and its production, distribution, and evolution paths, Friedlander introduced a conceptual framework for characterizing instruments used for aerosol measurements (Friedlander, 1970, 1971). Following his framework, the size distribution and number concentration of atmospheric aerosol particles are detected through electrostatic methods and condensational growth. The latter approach is the only technique available for detecting uncharged sub-100 nm particles. Consequently, it has become the dominant technique for assessing the integrated concentration of particles larger than a minimum size.

Since P. J. Coulter and J. Aitken published their observations dealing with the role of a fine airborne particle in the vapor condensation process in 1875 and 1880 separately (Spurny, 2000), the need to understand the phenomena has inspired the development of several particle counting instruments and led to various methods to quantify their performance under different operating conditions (Kangasluoma and Attoui, 2019; McMurry, 2000a; McMurry, 2000b). Several reviews have discussed the development of this technique in atmospheric aerosol measurements (Curtius, 2006; Kerminen et al., 2018; Kulmala et al., 2004; McMurry, 2000b). McMurry divided the history of condensation nucleus counters into two main sections through the end of the twentieth century – the development of expansion-type instruments and steady-flow condensation nucleus counters (McMurry,



2000a). Sem describes the designs of three commercial condensation particle counters (CPCs) and characterizes the particle diameter with 50% detection efficiencies of a TSI CPC (3025A, 3022A, and 3010) (Sem, 2002). Two recent comprehensive reviews by Kangasluoma et al. focus on developing instruments that can measure the particle size distribution down to the size of large molecules (Kangasluoma and Attoui, 2019; Kangasluoma et al., 2020). Kangasluoma et al. provide an in-depth review of the effort to advance the technology toward sub-10 nm size distribution measurements, summarize the current understanding of the characteristics of several systems, and identify instrumental limitations and potential advances for accuracy improvement in sub-3 nm particle counting.

Although most CPCs use butanol vapor to grow the aerosol particles, researchers notice that the working fluid plays a critical role in determining the size detection limits, as the vapor properties affect how the vapor condenses upon the particle to enlarge its size for detection. Stolzenburg and McMurry first described the effect of working fluid on size-dependent activation efficiencies with the laminar flow ultrafine condensation particle counter (Stolzenburg and McMurry, 1991), then theoretically described the effect of the working fluid. Experimental studies by Iida et al. (Iida et al., 2009) complemented their theory. Magnusson et al. concluded that working fluids with high surface tension (such as water and glycerol) could lead to a smaller activation size and reduce the lower size detection limit in the CPCs (Magnusson et al., 2003). As a working fluid, water avoids the health and safety concerns of butanol or other chemicals. Additionally, water-based CPCs reduce the requirement of chemical storage, maintenance effort, and ventilation system and eliminate water condensation and absorption into alcohol working fluids during operation in humid environments (Liu et al., 2006). Over the last few decades, mixing type water-based condensation systems have been used to capture particles for online chemical speciation instruments (Khlystov et al., 1995) and for high-flow condensation particle counting (Parsons and Mavliev, 2001).

The first laminar flow water-based condensation methods were introduced by Hering and Stolzenburg (2005), who introduced the concept of a warm, wet walled condenser for water-based condensational growth. The first implementation of this concept was the two-stage water CPC (Hering et al., 2005). The performance of several versions of this two-stage water-based CPC was intensively evaluated in the 21st century (Biswas et al., 2005; Hakala et al., 2013; Hering et al., 2005; Iida et al., 2008; Keller et al., 2013; Kupc et al., 2013; Kurten et al., 2005; Liu et al., 2006; Mordas et al., 2008; Petaja et al., 2006). In 2014, Hering and her co-workers further improved the laminar flow water CPC with a third stage to moderate the temperature profile between the growth tube and optics (Hering et al., 2014). This advanced design enables the capture of water vapor by the third moderator stage, such that conditions inside the instrument can be self-sustaining with regard to water consumption under moderate to high relative humidity (Hering et al., 2019; Kangasluoma and Attoui, 2019)). This feature has been commercialized in the "MAGIC" CPC (Moderated Aerosol Growth with Internal water Cycling, ADI). This concept also enables a higher temperature for the second 'initiator' stage, providing supersaturation can be created between the conditioner and initiator and leads to the activation of 1 nm particles without homogeneous water droplet formation (Hering et al., 2017; Kangasluoma and Attoui, 2019). It further allows flexibility in operating temperatures and a lower detection threshold, and hence has been named the "versatile" water CPC, or vWCPC. The vWCPC has been commercialized by TSI as the Model 3789

Many field deployments confirm that the water-based CPC has comparable performance to a butanol-based CPC when examining urban pollution and diesel combustion aerosol (Franklin et al., 2010; Jeong and Evans, 2009; Kaminsky et al., 2009; Keller et al., 2013; Lee et al., 2013; Sharma et al., 2011; Tsang et al., 2008). Based on these promising research results, it is desirable to explore the advanced water-based CPC for airborne measurements.

Airborne aerosol measurements provide researchers with *in situ* atmospheric properties across various spatial scales up to thousands of kilometers. However, it also creates design and characterization challenges due to the rapid change in environmental



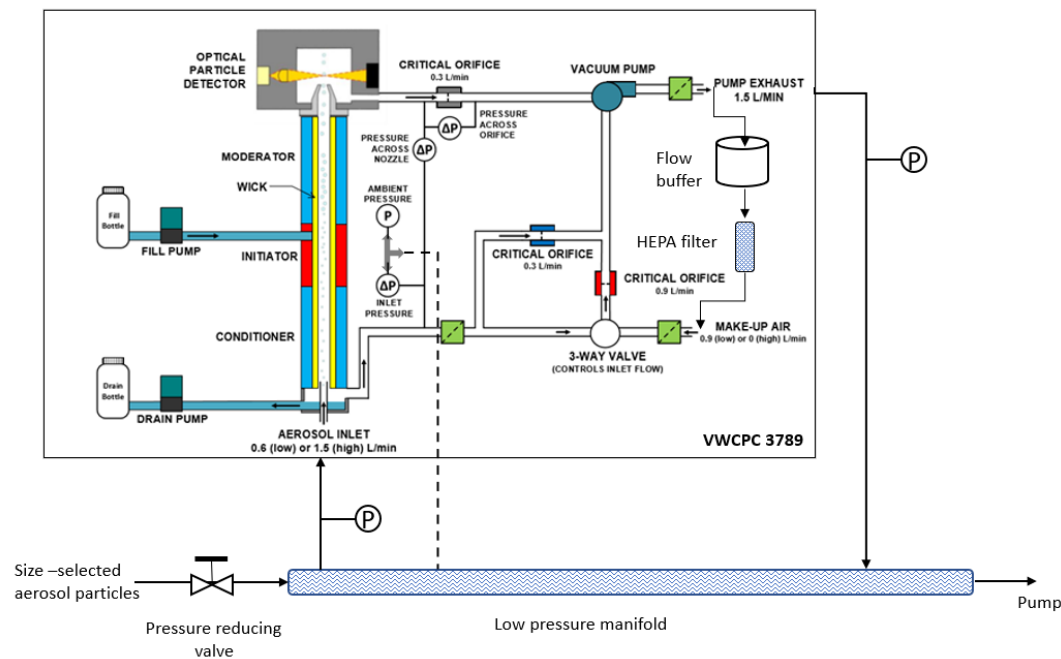
conditions. The pressure dependency of the counting efficiency of non-water-based CPCs has been explored in several studies (Hermann et al., 2005; Hermann and Wiedensohler, 2001; Seifert et al., 2004; Weigel et al., 2009). However, our literature research shows no records about the water-based CPC being evaluated under low-pressure conditions. Thus, this study characterizes a versatile vWCPC and its counting efficiency changes under such low-pressure conditions. Measurements and modeling were done over the pressure range from 500 hPa to 1000 hPa. We examined the 7-nm configuration rather than the 2-nm configuration because the equilibrium water vapor pressure at the maximum temperature in the 2-nm configuration exceeds 500 hPa. A three-stage operating temperature profile simulation was carried out to understand the supersaturation profile inside the water-based condensational growth tube and guided the optimization of the operation setting. A previously developed particle growth model was used to evaluate the pressure change effect on aerosol particle activation and droplet growth. A simplified theoretical analysis is presented to evaluate effects associated with high particle concentrations. Data at low pressures were obtained using a mono-dispersed aerosol of various chemical compositions compared to a CPC operated at atmospheric pressure and a parallel aerosol electrometer at low pressure.

## 2 Materials and Methods

### 2.1 Instrument modification

The vWCPC 3789 tested in this study uses a three-stage growth tube, as described by Hering et al. (2017). A single tube with a 6.3mm ID is lined with a wet, porous wick. It has three temperature regions, referred to as the conditioner, initiator, and moderator, with lengths of 73 mm, 30 mm, and 73 mm, respectively. The aerosol flow is 0.3 L/min. For the 7-nm configuration tested here, the factory temperature settings of the walls of the conditioner, initiator, and moderator regions are:  $T_{\text{cond}}=30^{\circ}\text{C}$ ,  $T_{\text{ini}}=59^{\circ}\text{C}$ , and  $T_{\text{mod}}=10^{\circ}\text{C}$ .

Several modifications were made to the unit in this study because commercially available vWCPC are not designed for low-pressure applications, as shown in Fig. 1. First, each vWCPC was tested to ensure it is vacuum-tight. Therefore, the make-up flow port and exhaust port were blocked during the vacuum-tight check. This step guarantees the instrument operates appropriately under conditions of a significant positive pressure difference between ambient and internal pressures, which mimic the characteristic of the high-altitude operation on an aircraft with a pressurized cabin. Secondly, the vWCPC is monitoring the inlet pressure, orifice pressure and nozzle pressure during the operation. Thus, we connected the ambient pressure port and inlet pressure port to the low-pressure manifold, which prevented triggering the warning and error indicator. Thirdly, we added a pressure transducer in the vWCPC inlet line and a pressure transducer (both Baratron 722B, MKS Instruments. Inc., Andover, MA, USA) in the exhaust line. Finally, when we operated with 1.5 lpm inlet aerosol flow, we blocked the make-up flow port. When we operated with 0.6 lpm inlet aerosol flow, 0.9 lpm flow from the exhaust line was filtered, passed through a flow buffer, and then made up the 1.5 lpm vacuum flow. Note that under both operating conditions, the aerosol flow passing the condensation tubing and optical particle detector is 0.3 lpm.



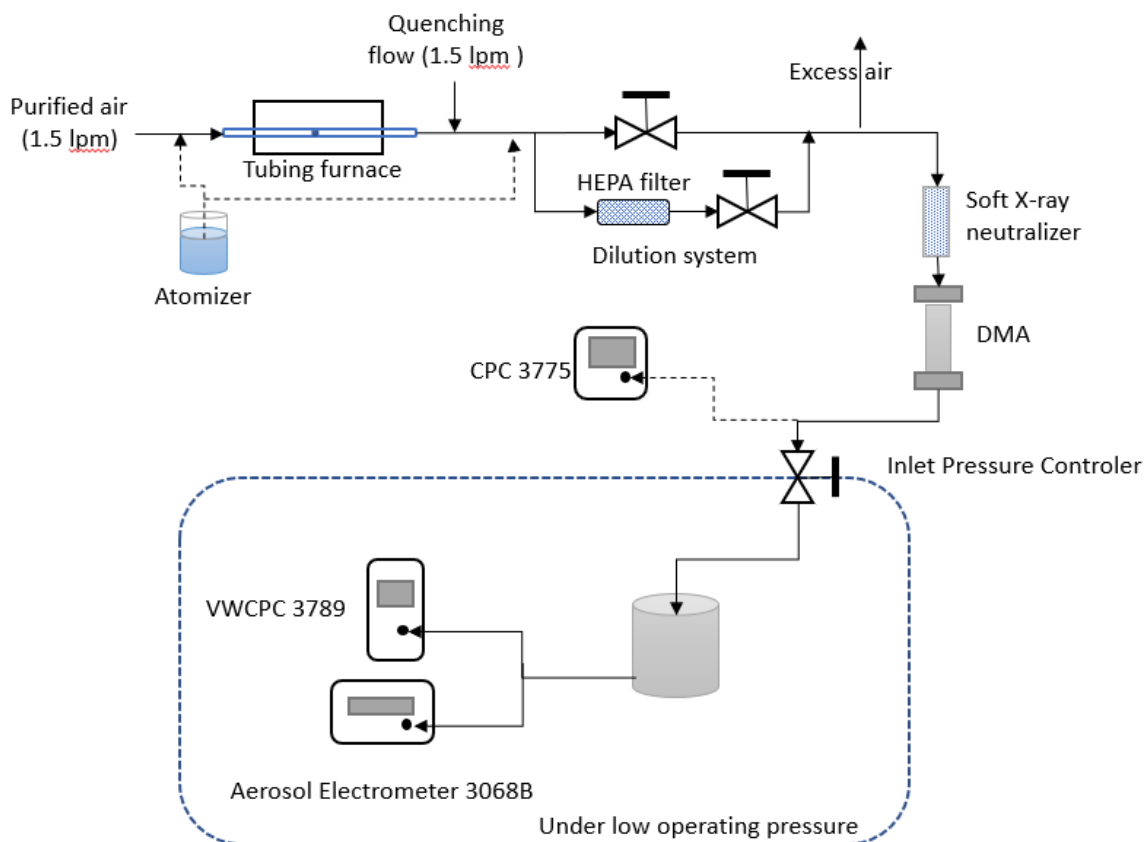
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**Figure 1: Schematic of the modified vWCPC sensors and flow system (TSI Incorporated, 2019).**

## 2.2 Experimental characterization setup

The low-pressure calibration setup of the vWCPC is shown in Fig. 2. Polydisperse ammonium sulfate (AS) aerosols were produced in a tube furnace generator (Lindberg/Blue). Polystyrene latex (PSL) particles were generated through an atomizer. After passing a dilution system, the aerosol particles were size-selected by a differential mobility analyzer (DMA, TSI 3081) using a soft x-ray neutralizer (TSI, advanced aerosol neutralizer 3088). The operating pressure was reduced by a constant pressure inlet (DMT) to simulate different flight level pressures (500 – 1000 hPa). For the counting efficiency determination, two reference sensors were used. One reference sensor was a CPC 3775 (TSI), which was connected directly to the monodisperse flow after the DMA and operated at atmospheric pressure. Its inlet aerosol flow rate was 0.3 lpm. The other sensor, an aerosol electrometer (A.E. 3068B, TSI), was operated parallel with the CPC 3789 under low-pressure conditions. Both CPC 3789 and A.E. were run at 0.6 lpm inlet flow to ensure equal diffusive particle loss in the aerosol pathway. The DMA's sheath flow was typically 20 lpm, resulting in a sheath to aerosol flow ratio and a non-diffusive mobility resolution of 13.

The CPC counting efficiency is defined by the ratio of the particle number concentration measured by the CPC 3789 and the reference particle number concentration measured by the A.E. Multiple charged particles induce a reading error in the reference particle number concentration. This error is negligible for particle sizes less than 70 nm compared to other experimental errors. (Hermann, 2000; Hermann, 2001). For aerosol particles larger than 70 nm, an empirical correction was estimated using the size distribution of the generated aerosol and the aerosol charging distribution (Tigges, 2015, "Bipolar charge distribution of a soft x-ray diffusion charger") and the particle loss through the constant pressure inlet, which can be estimated based on the total number concentration difference measured by the CPC 3775 and vWCPC 3789.



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**Figure 2. Schematic of the vWCPC sensors and flow system under the low-pressure testing.**

Ammonium sulfate was the primary material for this study and was dissolved into deionized water for aerosol generation using atomization techniques. The water-insoluble chemicals, such as humic acid and oleic acid, were atomized from a water suspension after at least 10 minutes of ultrasonic aided mixing. The properties of the other tested aerosol particles were included in Table S1.

### 135 2.3 Numerical simulation

The numeric modeling approach used here has been documented in previous publications (Hering et al., 2014; Lewis and Hering, 2013). First, the temperature and humidity profiles were computed using the finite element modeling software - COMSOL Multiphysics®. The configuration in Hering's research consists of a 4.6 mm I.D. tube extending through a 154 mm conditioner, a 76 mm initiator, and a 100 mm moderator, with a design flow rate of 1.5 L/min. Next, the particle growth, temperature and humidity profiles were calculated using a numerical model developed by Lewis and Hering (Lewis and Hering, 2013), written in Igor Pro (Wavementrics, Beaverton, OR). Their simulation results suggest that the three-stage configuration is superior in decreasing the amount of water vapor and lowering the particle loss and variation in detection and collection, avoiding the side-effect of heating the flow. With this "moderated" approach, a short, warm, wet-walled initiator provides sufficient water vapor for activation, followed by a cool-walled moderator for particle growth. A recent simulation study (Bian et al., 2020) confirmed Hering's finding with different temperature settings.

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In general, enhancing the temperature difference between the initiator and the conditioner can obtain higher supersaturation and smaller activation size. Furthermore, shifting the 70 °C temperature difference window by decreasing the conditioner temperature



(from 9 °C to 1 °C) further reduces the activation size. In addition, Hering's paper pointed out that the final droplet size decreases from around 4.5 μm to 2.5 μm with the increase of the aerosol inlet flow rate from 0.4 lpm to 1.5 lpm. This behavior is consistent with Bian's result, which shows that the flow rate varied by a factor of 2.5, the final size decreased by 43%.

Numerical simulations of the temperature and humidity profiles and the particle growth were calculated for the geometry of the vWCPC 3789 described above, using the methods published by Hering's group (Hering et al., 2014; Lewis and Hering, 2013). The assumption and equations used for the simulation are included in the supplemental document. More details are discussed in section 3.2.

### 3 Results

#### 3.1 Pressure dependence of the vWCPC counting efficiency

Using the low-pressure testing setup shown in Fig. 2, the counting efficiency of a CPC 3789 was measured between 500 hPa to 920 hPa for particles of 15 nm, 25 nm, and 100 nm. The aerosol concentrations in this test were maintained  $2\sim 4 \times 10^4 \text{ cm}^{-3}$ . The obtained counting efficiencies for the manufacturer's 7 nm cut-off setting are shown in Fig. 3. With the 7 nm cut-off size manufacturer setting, the conditioner, initiator, and moderator temperatures were 30 °C, 59 °C, and 10 °C.

During the testing, the temperature variations in the conditioner and moderator were less than  $\pm 0.5 \text{ °C}$ . For the "7 nm" temperature setting, the initiator temperature has a variation of  $\pm 1 \text{ °C}$ . The y-axis error bar indicates the standard deviation of the counting efficiency averaged over ~5 minutes of sampling time. Fig. 3 shows that the counting efficiencies decrease with the decrease of the operating pressure. The decreases become significant and larger than 10% when the operating pressure was lower than 500 hPa.

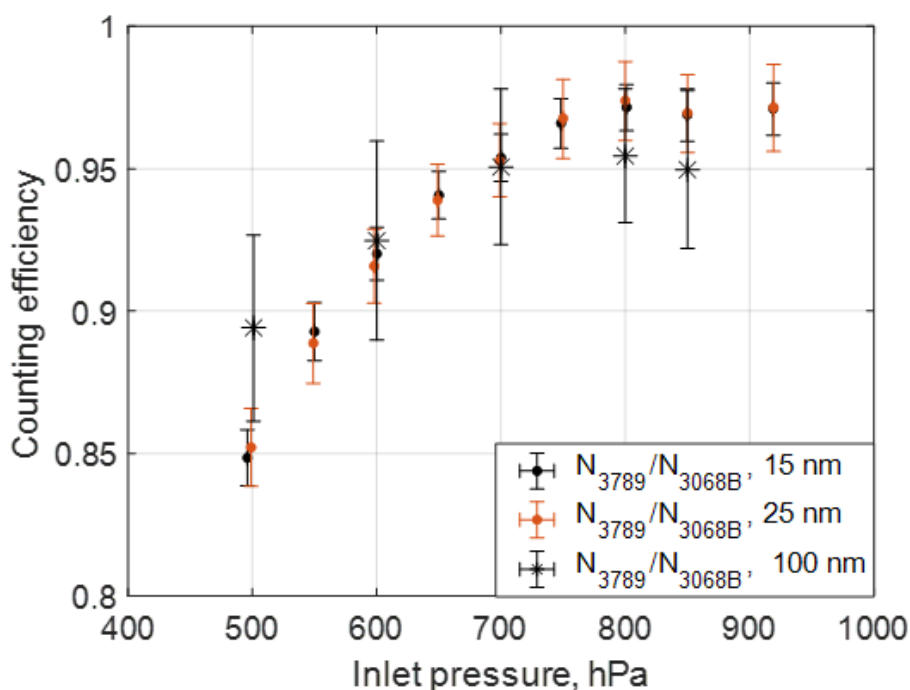


Fig. 3, CPC 3789 counting efficiency as a function of the inlet operation pressure at  $T_{\text{cond}} = 30 \text{ °C}$ ,  $T_{\text{ini}} = 59 \text{ °C}$ , and  $T_{\text{mod}} = 10 \text{ °C}$ .  $N_{3789}/N_{3068B}$  is the total number concentration ratio between the vWCPC and the electrometer.



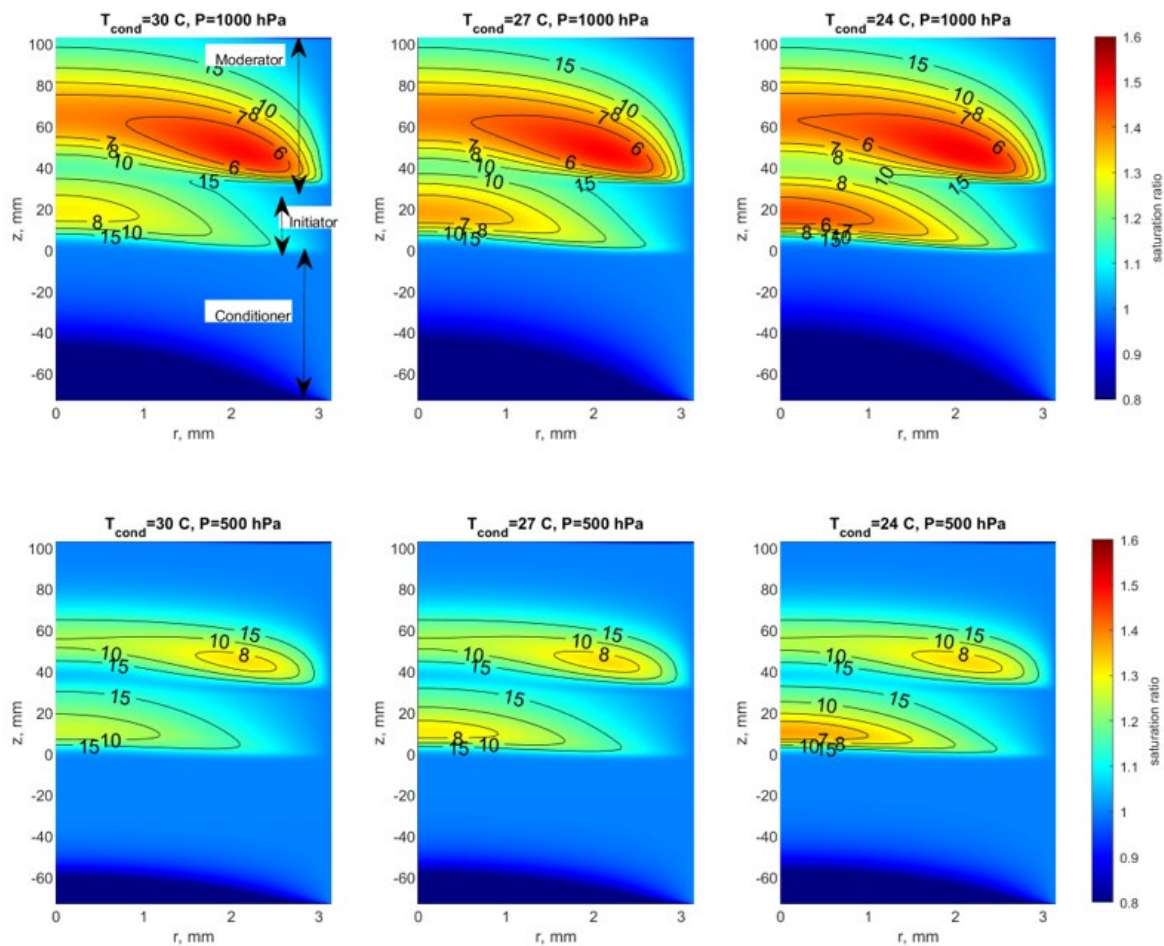
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Based on the  $\kappa$ -Köhler theory and the supersaturation (relative humidity-1) estimated at the centerline of a three-stage chamber in Fig. S2, ammonium sulfate particles larger than 9.7 nm should all be activated when the saturation ratio is larger than 1.05, even if we assume that the supersaturation profile near the wall has an 85% drop from the centerline condition. Thus, the low counting efficiency under the low-pressure operation condition is not limited by aerosol activation.

175 Previous simulation studies show that the centerline saturation ratio is not sensitive to the wall temperature of the moderator (Bian et al., 2020; Hering et al., 2014). However, under the low-pressure condition (e.g., 500 hPa), the saturation profile peaked earlier but lower than the saturation profile under the standard condition (1000 hPa), as shown in Fig. S2. Also, decreasing the conditioner temperature (while maintaining the same temperature difference between the initiator and the conditioner) provided higher saturation ratios in the initiator and more water vapor for particle growth, which is also consistent with the previous growth tubing  
180 simulation (Bian et al., 2020). Thus, in the following section, we examine the temperature effect on the vWCPC performance under the low-pressure condition. Furthermore, with the aid of the simulation, we study how to overcome the counting efficiency decrease experimentally.

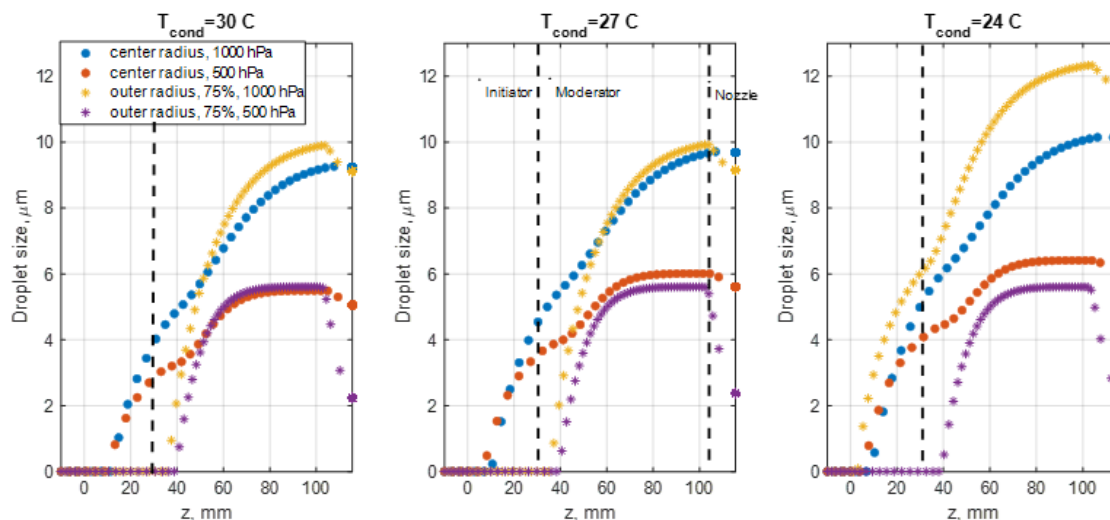
### 3.2 Modelled pressure dependence of the vWCPC counting efficiency at different operating temperatures.

The simulated saturation profiles of a three-stage growth tube at different operation pressure (1000 and 500 hPa) for three different  
185 conditioner temperatures (30 °C, 27 °C, and 24 °C), all with initiator temperature of 59°C and moderator temperature of 10°C, are presented in Fig.4. The predicted droplet size evolution along the growth tube of this vWCPC at different operation pressures (1000 and 500 hPa) under three conditioner temperatures (30 °C, 27 °C, and 24 °C) is included in Fig. 5. In most water-based CPCs, the aerosol particle growth was initiated by the temperature rise from the conditioner to the initiator. The growth rate was modulated  
190 by the temperature decrease from the initiator and the moderator. The maximum saturation ratio calculated at the centerline usually occurs downstream of the initiator exit due to the water vapor diffusion delay. However, in Fig. 4, we observed a double-peaked saturation ratio profile appeared for three different conditioner temperature settings under both operation pressures (1000 and 500 hPa). The maximum saturation peak occurs inside the moderator under most simulated conditions, except when the operation pressure decreased to 500 hPa at the conditioner temperature of 24 °C, the highest saturation peak occurs inside the initiator. Under the standard pressure, this configuration has the advantage that the peak extends close to the wall, resulting in high counting  
195 efficiency and a sharp lower cut-off size, as shown in Fig. 5. With the decrease of the operating pressure, the saturation peaks showed a substantial reduction, which was also associated with both, the lower cut-off size increasing in Fig. 4. and the growing droplet size decreasing in Fig 5. When the conditioner temperature is 27 °C or 30 °C, with decreasing of the operating pressure from 1000 hPa to 500 hPa, one 8 nm seed particle grew to a smaller size (~40% reduction in the droplet diameter), no matter the particle entered the growth tube in the centerline or near the wall. The seed particles entering the centerline of a growth tube got  
200 activated in the initiator. Delay of the activation occurred for the seed particles entering the growth tube away from the centerline, and those particles at 75% of radius started growing in the moderator.



205 Fig 4. Simulation of the CPC3789 saturation profiles at 1000 hPa (upper panel) and 500 hPa (bottom panel) under the different conditioner temperatures (30 °C, 27 °C, and 24 °C) with the initiator temperature is 59 °C, and the moderator temperature is 10 °C. The color bar indicates the humidity (the saturation ratio) change inside the three-stage growth tube. The contour line indicates the saturation ratio necessary to activate 6, 7, 8, 10, 15 nm seed particles.  $r$  is the distance from the centerline.  $z$  is the distance from the entrance of the initiator, which means that the conditioner is from -70 to 0 mm.





210 **Fig. 5.** Predicted droplet size evolution along the growth tube (at centerline or 75% of the inner tubing radius) of the CPC 3789 under the different conditioner temperatures (30 °C, 27 °C, and 24 °C) with the initiator temperature is 59 °C, and the moderator temperature is 10 °C. Starting particle size 8 nm.

Guided by the above analysis and the observations, we maintained the temperature settings in the initiator at 59°C and moderator  
215 at 10°C and varied the conditioner temperatures by 3 °C. The aerosol particle concentrations were maintained at around  
 $6 \times 10^3(\text{cm}^{-3})$  in this study to avoid the concentration effects (more discussion in section 3.3). Under each conditioner temperature,  
we examined the impact of the operating pressure on the aerosol particle growth inside the three-stage growth tube through the  
simulation (shown in Fig. 4, 5, and S2) and on the vWCPC counting efficiency change through the experiment (as shown in Fig.  
6). When the conditioner temperature was set at 30 °C or 33 °C, the counting efficiency drops as the inlet pressure is lowered from  
220 1000 hPa to 500 hPa. However, when the conditioner temperature was decreased to 27 °C or 24 °C, we did not observe a noticeable  
decrease in the counting efficiency with the operating pressure decrease.

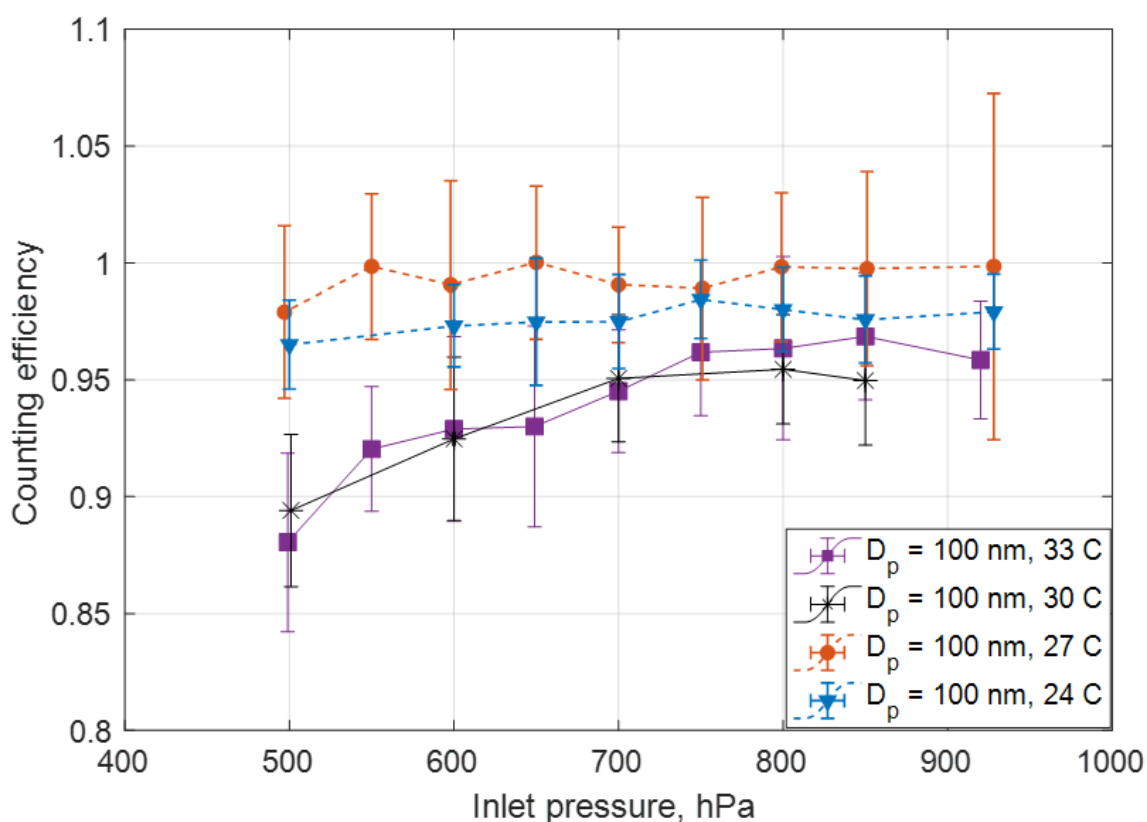


Fig. 6, CPC 3789 counting efficiency as a function of the inlet operation pressure at four conditioner temperatures of  $T_{\text{cond}} = 24, 27, 30,$   
225  $33\text{ °C}$  ( $T_{\text{ini}}=59\text{ °C}$ ,  $T_{\text{mod}}=10\text{ °C}$ ).

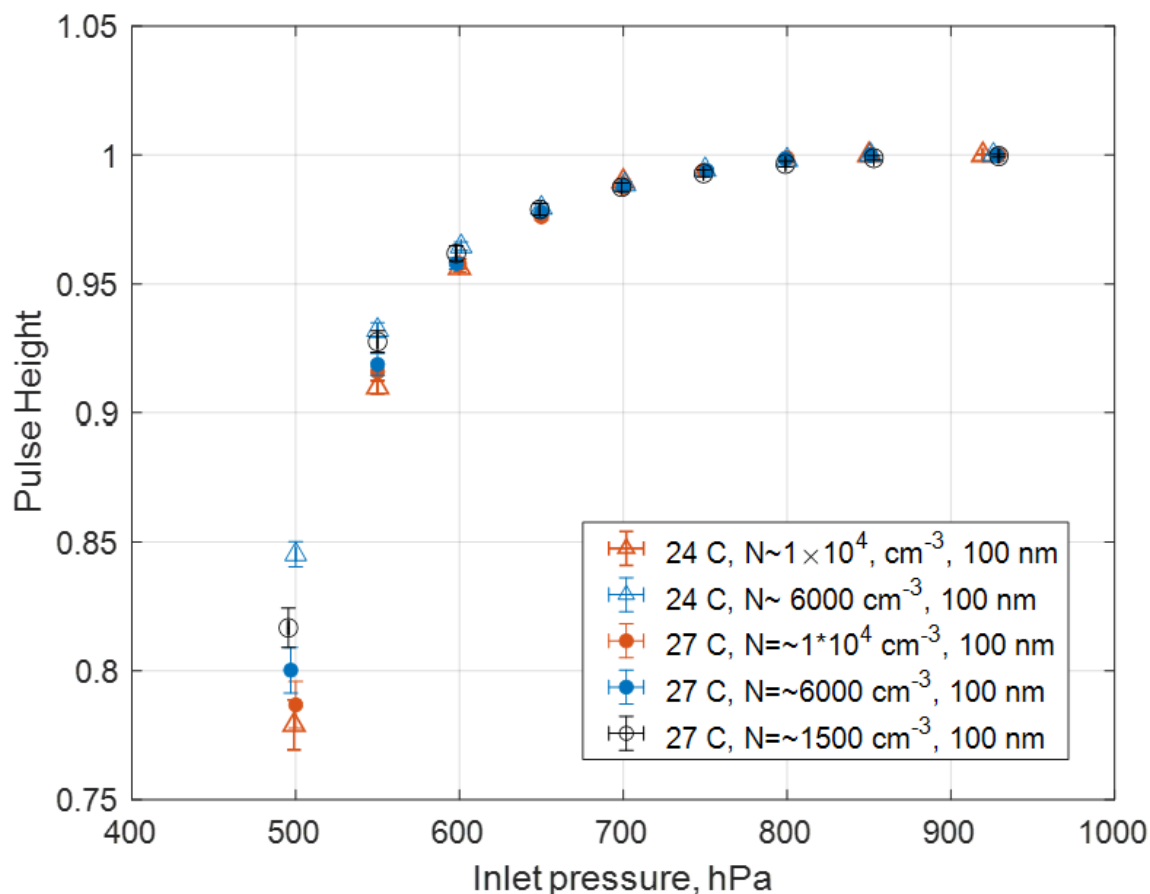
The experimental results suggest that increasing the temperature difference between the conditioner and the imitator affected the vWCPC 3789 counting efficiency. As shown in Fig. 6, the counting efficiencies of the  $T_{\text{cond}}=27\text{ °C}$  setting were maintained close to 1 when the operating pressure dropped to 500 hPa. This observation suggests that the low counting efficiency observed in Fig. 3 was not mainly caused by the particle loss inside of the instruments. Fig. 5 suggests that although the 2<sup>nd</sup> saturation peak in the moderator activated the seed particles and is capable of maintaining droplet growth, about 10% of particles may not grow large enough to be detected in the optical chamber. Therefore, the counting efficiency is more susceptible to the 1<sup>st</sup> peak of the supersaturation profile in the initiator. In addition, the absolute saturation also set the threshold for successfully operating this vWCPC 3789 under lower pressure. Combining the observations from Fig 6 and the simulation in Fig. 4 and 5, when the simulated saturation is over 1.3, the counting efficiency maintained close to 1 for aerosol particles larger than 15 nm under low-pressure conditions. We also noticed that the counting efficiency curve is slightly lower when operating the conditioner at 24 °C than the conditioner was set to 27 °C. That is possibly due to the high supersaturation profile inside the three-stage tube, leading to larger droplets, especially close to the wall and increased loss at the tubing wall and through the focusing nozzle.

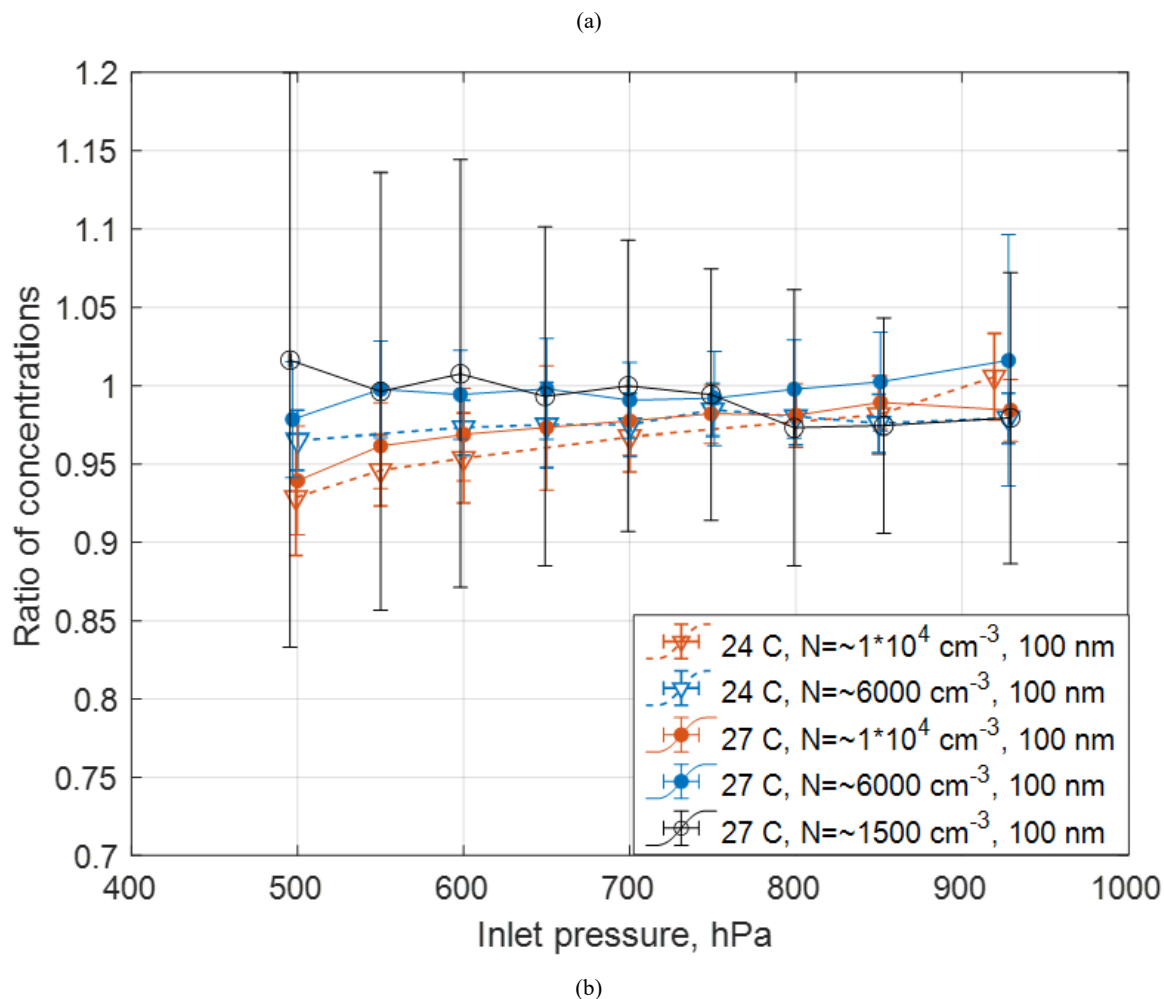
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### 3.3 Particle concentration and pressure effects on droplet size and vWCPC counting efficiency

240 In typical operation, the manufacturer reports that the single-particle counting is maintained up to a concentration of  $2 \times 10^5$  ( $\text{cm}^{-3}$ ). The pulse height is above 90% for moderate concentrations ( $\sim 10$ - $5,000 \text{ cm}^{-3}$ ). This vWCPC uses pulse height to indicate the fraction of the particle population generating an acceptably high pulse. Thus, we examined if this concentration threshold would hold under low-pressure conditions. For 100 nm particles, we observed that the pulse height decreases with the decrease of the operating pressure. When the operation pressure decreased from 550 hPa to 500 hPa, the pulse height decreased from above 90% to around 80%, as shown in Fig 7 (a). Meanwhile, the measured 10% reduction threshold ( $N_{\text{meas}}$ , under 500 hPa) with the pulse height values for 100 nm aerosol particles is about  $1 \times 10^4$  ( $\text{cm}^{-3}$ ), as shown in Fig. 7 (b). When the aerosol concentration is larger than  $2 \times 10^4$  ( $\text{cm}^{-3}$ ), decreasing the pressure affected the measured aerosol concentration more significantly, as shown in Fig. S5(b). Comparing the pulse height curves, while the conditioner temperature operated at 24 °C and 30 °C, Fig. S5(a) showed that the 30 °C case suffered more water vapor shortage while decreasing the operating pressure. Additionally, there is no significant difference between the measured 10% reduction threshold between 20 nm and 100 nm particles, when the particle concentration is less than  $1 \times 10^4$  ( $\text{cm}^{-3}$ ). This observation is consistent with the simulation estimated 10% reduction of  $s$  and  $D_p$  happened when the  $N_{7\mu\text{m}} \sim 8.5 \times 10^3$  ( $\text{cm}^{-3}$ ) as shown in the supplemental material. Moreover, it indicates that we can monitor the pulse height value to detect the undercounting issue. When the pulse height is less than 80%, the measured aerosol concentration by vWCPC 3789 is about 10% less than the aerosol concentration measured by the electrometer.





260 Fig. 7. The water depletion due to the aerosol number concentration, illustrated by (a) the pulse height generated in the optical detector,  
265 (b) the counting efficiency as a function of the inlet pressure. Results are shown with the conditioner temperatures were set at 24 °C and  
270 27 °C.

### 3.4 Chemical composition effect on the vWCPC counting efficiency

265 We examine the counting efficiency of the CPC 3789 using aerosol particles with different chemical compositions and water  
solubility, as shown in Table S1. We choose three types of 100 nm aerosol particles: water-insoluble particles, such as oleic acid  
particles and PSL, and highly hydrophilic ammonium sulfate particles. Running the 100 nm particles under different operating  
pressures, we get similar counting efficiencies. However, the counting efficiencies of the oleic acid and PSL particles showed more  
significant uncertainty than that of the ammonium sulfate particles. Therefore, to understand the condensation effect on the droplet  
270 size inside the growth tubing, we proceed with a simple analysis to determine the impact.

The chemical composition also affects the observed counting efficiencies. During this test, 100 nm aerosol particles were atomized  
from the ammonium sulfate, sucrose, humic acid and PSL solutions or suspensions, as shown in Fig. 8. Note that we could not



characterize the oleic acid particles because oleic acid particles evaporated under low-pressure conditions and caused a significant variation in the number concentration and size distribution of size-selected particles. We noticed that the PSL particle curve has more substantial variation comparing to the other aerosol particles. The particle surface is very hydrophobic, and the droplet growth process will be affected by the remaining water or surfactant on the particle surface. The counting efficiency of humic acid particles was slightly lower than the counting efficiency of AS aerosol particles, which can likely be explained by the absorbing properties of humic acid. The counting efficiency of sucrose is significantly lower than the counting efficiency of AS (10% lower). This observation is consistent with the previous study (Hering et al., 2017).

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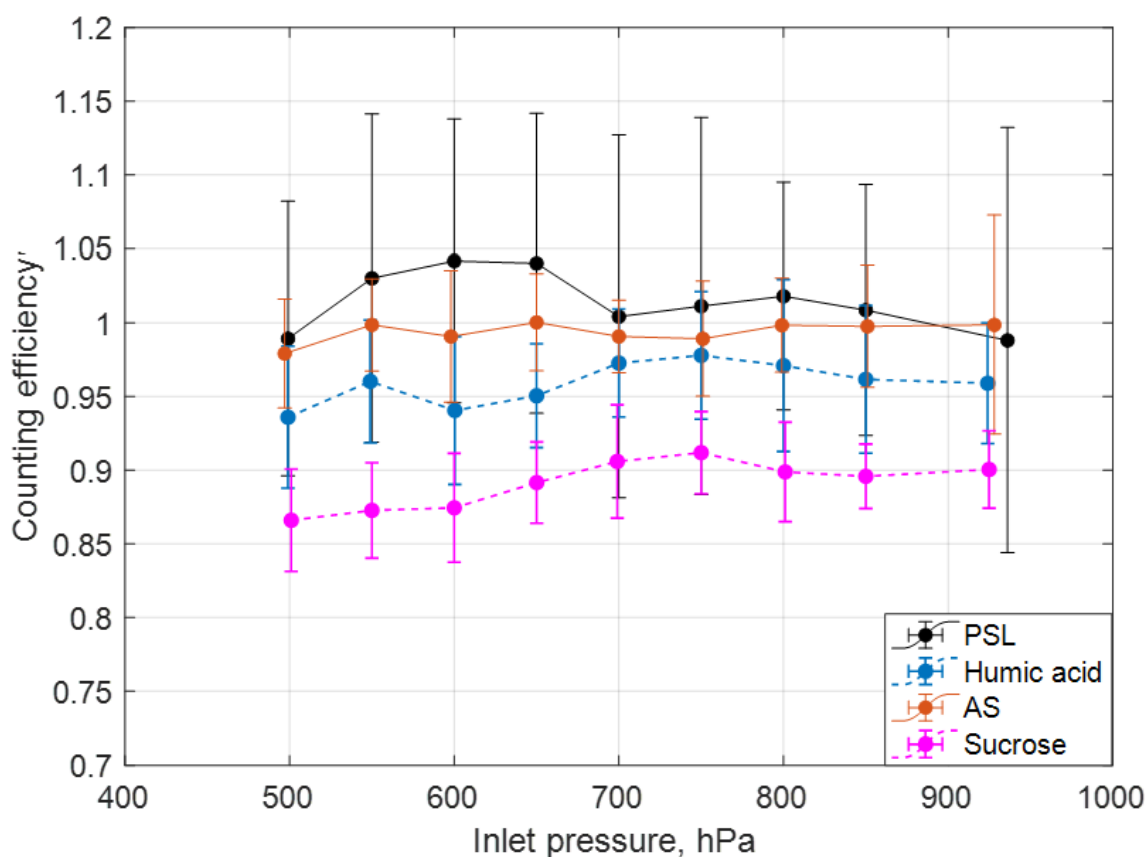


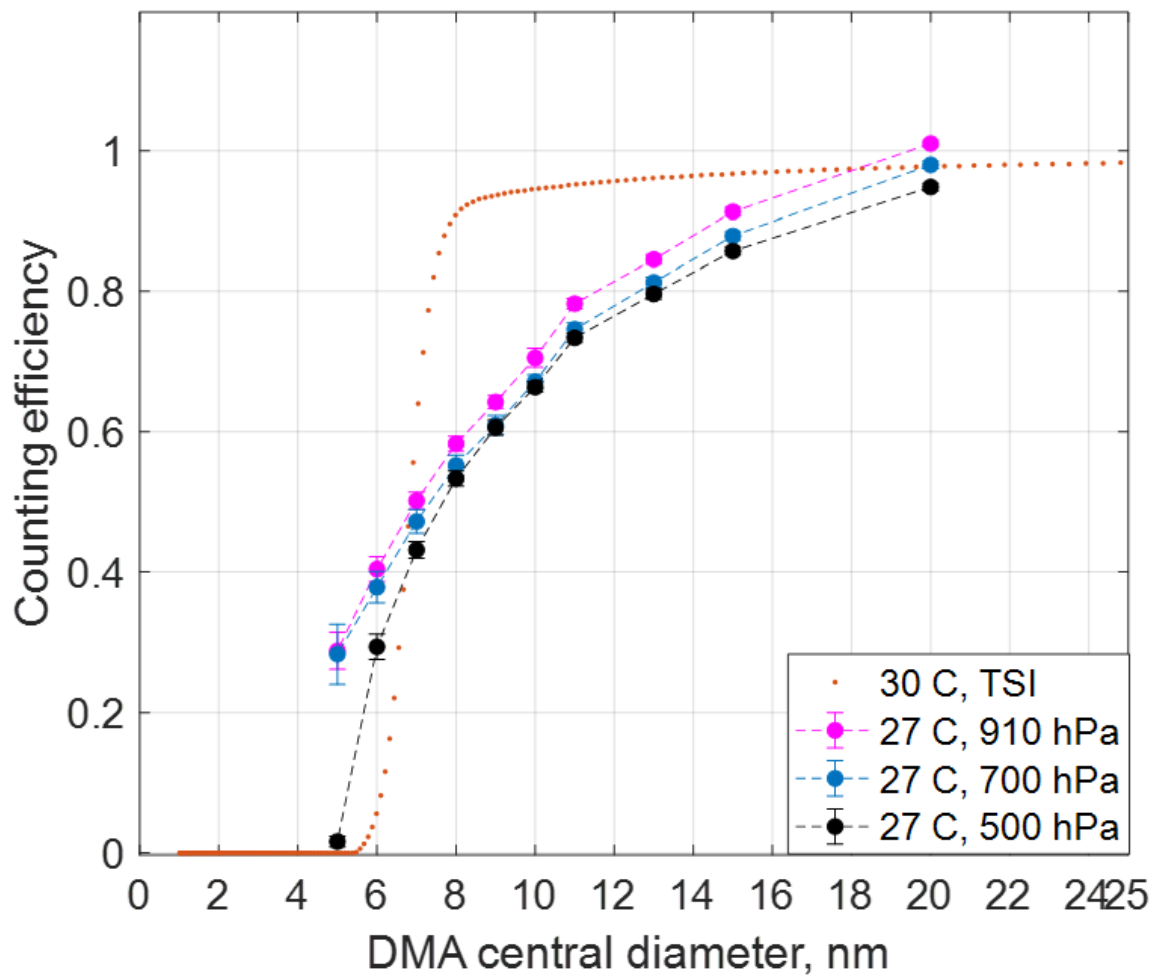
Fig. 8. CPC 3789 counting efficiency at different operating pressure with four different types of aerosol particles, when the temperature conditions are  $T_{\text{cond}} = 27\text{ }^{\circ}\text{C}$  and  $T_{\text{ini}} = 59\text{ }^{\circ}\text{C}$ .

### 3.5 Temperature dependence of the vWCPC cut-off size

285 For the butanol-based CPC, the CPC cut-off size was strongly influenced by the temperature difference between the saturator and condenser. Typically, with the increase of the temperature difference, the cut-off size decreases. We observed that this trend held well for the water CPC. The temperature effect on the counting efficiency of the CPC 3789 under low-pressure conditions was presented in Fig. 9. With the decrease of the conditioner temperature, the temperature difference between the initiator and the conditioner increases. As expected, the cut-off size slightly moved to lower than 7 nm. However, with the decrease of the operating  
290 pressure, the cut-off size increased slightly. Under two conditioner temperatures ( $24\text{ }^{\circ}\text{C}$  and  $27\text{ }^{\circ}\text{C}$ ), the cut-off size was maintained

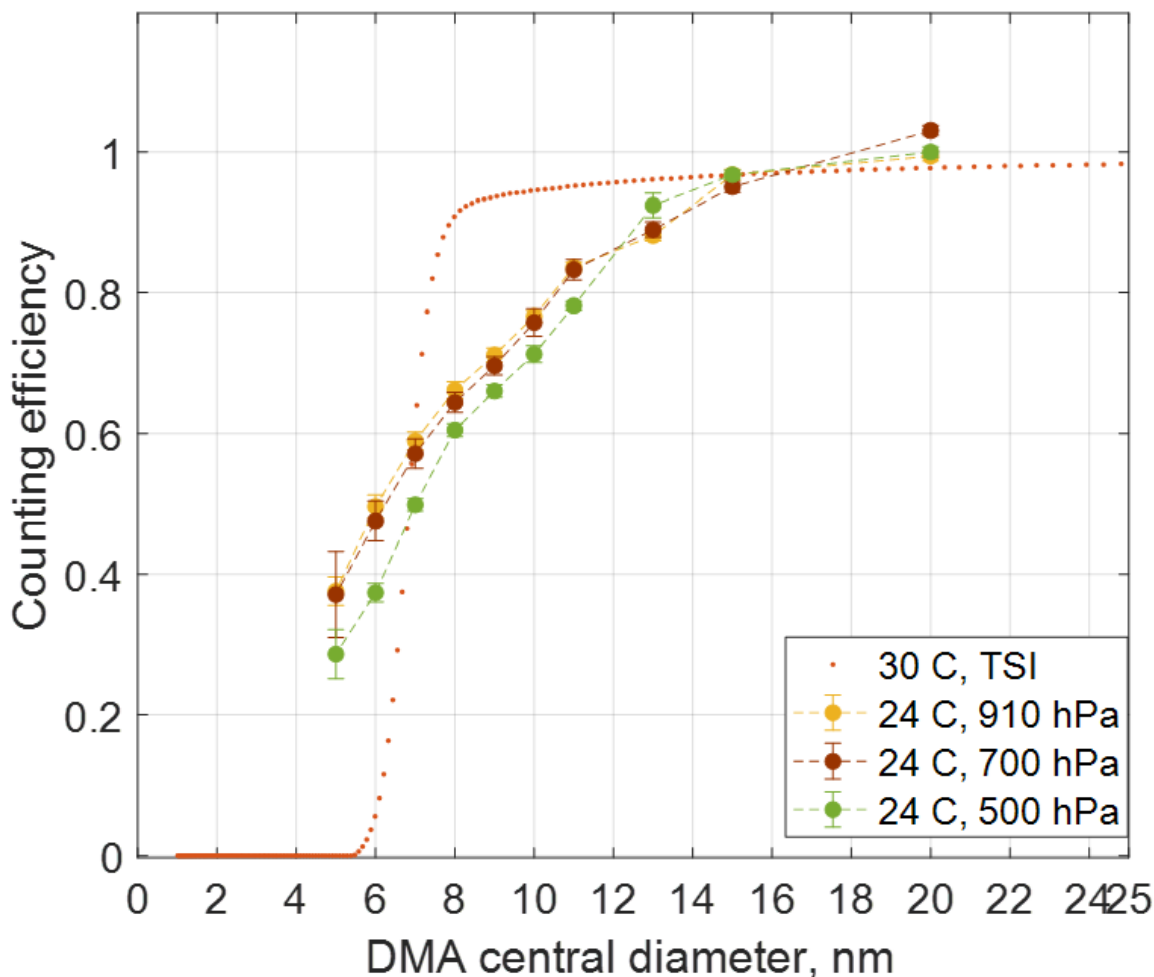


between 6-8 nm, even with the pressure dropped to 500 hPa. Thus, both settings are suitable for airborne operations up to 5.5 km with a 7 nm cut-off size.



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(a)



(b)

Fig. 9. CPC 3789 counting efficiency changes as a function of the different operating pressures, when the initiator temperature is 59 °C, and the moderator temperature is 10 °C, at two different conditioner temperatures (a)  $T_{\text{cond}} = 24$ , (b)  $T_{\text{cond}} = 27$  °C.

#### 300 4 Conclusions

This study discusses the modification and characterization of the versatile water CPC (TSI 3789) operating under low-pressure conditions. A commercially available vWCPC was modified to report the environmental pressure during the airborne operation. A calibration setup was built to study the vWCPC counting efficiency as a function of operating pressure (500 – 920 hPa) and different conditioner temperatures (24 – 33 °C) at factory settings for the initiator and moderator temperatures (59° and 10°). The  
305 vWCPC with the manufacturer settings worked as expected under the standard ambient condition. Under low-pressure conditions, we observed that the counting efficiency of this vWCPC at the factory settings decreased with the decrease of the operating pressure. Although the numerical simulation with the current assumption could not fully explain the above phenomena, the numerical simulation and the dimensionless analysis results showed that the saturation peak under 500 hPa was lower than the saturation peak under 1000 hPa but arose closer to the entrance of the initiator. Besides, at the same operation pressure, with the



310 conditioner temperature decreased, the saturation peaks increased. Aided by the simulation results, we examined the conditioner  
temperature effect on the counting efficiency. We noticed that setting the conditioner temperature to 27°C was optimal for operating  
the vWCPC under various pressure conditions. However, we also observed that the vWCPC's counting efficiency curve varied  
based on the aerosol total number concentration under higher conditioner temperatures. The simplified water depletion estimation  
suggested that the 10% reduction of  $s$  and  $D_p$  happened when the  $N_{7\mu\text{m}} \sim 8.5 \times 10^3 (\text{cm}^{-3})$ . We measured the pulse height reported  
315 by the vWCPC with different aerosol total number concentrations. We observed a similar decreasing trend as we decrease the  
operating pressure. When the pulse height was larger than 80%, and the particle concentration is less than  $1 \times 10^4 (\text{cm}^{-3})$ , 20 nm  
and 100 nm experienced around 10% reduction in the measured concentration. The chemical composition of aerosol also  
contributes about 20% uncertainty in the counting efficiency of CPC, and there is no significant trend associated with the operating  
pressure changes.

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#### Data availability

The CPC data in the study are available upon reasonable request to Fan Mei ([fan.mei@pnnl.gov](mailto:fan.mei@pnnl.gov)).

#### Author contributions

330 F.M., M.S.P., S.H., and J.W. designed the research. F.M. carried out the measurements. F.M. led the analyses, and S.S. led the simulation. F.M.  
led the writing, with significant input from S.H. and J.W. and further input from all other authors. B.S. and J.T. acquired the financial support  
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#### Competing interests

The authors declare that they have no conflict of interest.

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