

1. Materials and Methods

1.1 Droplet Growth Efficiency

We measured the droplet growth efficiency, which refers to the initiation of vapor condensation on the surface of the aerosol particles acting as nuclei and the growth, achieved through the TCALC as a function of the temperature of the vapor flow generated through the saturator (T_{sat}), the temperature of the growth tube wall (T_{gt}), the aerodynamic diameter of the particles (d_p) and the relative humidity of the aerosol stream (RH). The experimental set up used is shown in Figure 1.

Sodium chloride aerosol was generated through a jet nebulizer (Salter 8900 Series; Salter Labs, Arvin CA), and was subsequently dried using a diffusion dryer (model 3062, TSI Inc., Shoreview MN), containing silica gel desiccant. Near-monodisperse aerosol of desired aerodynamic diameter ($d_p = 25, 50, 75, 100, 150, 200, 250$ and 300 nm), was obtained using an Aerodynamic Aerosol Classifier (AAC; Cambustion Ltd, Cambridge, United Kingdom) operating at 1.5 L min^{-1} .

Upstream of the mixing region of the TCALC, two humidifiers (MH-110-12F-4; Perma Pure LLC, Lakewood NJ) were placed: one was placed downstream of the AAC and the second was placed in parallel with a valve, controlling the relative humidity of the dilution air and subsequently of the aerosol flow. The aerosol flow was introduced in the mixing region at a flowrate (Q_a) of 8, 9 or 10 L min^{-1} , an ambient temperature (T_a) of 22°C , and an RH in the range of 9 – 60%.

Water vapor is generated through the saturator at a fixed temperature of 70, 75, 80 and 85°C . For these saturator temperatures, we have calculated the saturation ratio (S_R) and Kelvin

diameter (seed particle diameter that neither grows nor evaporates; d_p^*) as a function of the ratio of vapor-to-aerosol flowrate. Figure 2 shows that different saturator temperatures required different vapor-to-aerosol flowrate ratios (Q_v/Q_a), to reach maximum S_R or minimum d_p^* . Therefore, when the aerosol flow temperature (T_a) is 20 °C, a vapor-to-aerosol flowrate ratio of 0.25, 0.23, 0.18 and 0.13 should be used for a T_{sat} of 70, 75, 80, and 85 °C, to achieve S_R of approximately 1.9, 2.1, 2.3 and 2.5, respectively (Zervaki et al. 2024). Particle-free air was introduced into the saturator, and a vapor-saturated flow was produced at a controlled flowrate (Q_v) of 1, 1.2, 1.3, 1.4, 1.6, 1.8, 2, 2.3 or 2.5 L min⁻¹.

Following the mixing of the cold aerosol flow with the hot vapor-saturated flow, the mixed flow is introduced into the droplet growth region, which was further “actively” cooled using thermoelectric coolers. The temperature of the growth tube was controlled at 0, 5, 10, and 15 °C. We also evaluated the collector when no cooling was applied on the droplet growth region.

A Condensation Particle Counter (model UWCPC 3786; TSI Inc., Shoreview MN) was used along with an Optical Particle Sizer (model OPS 3330; TSI Inc., Shoreview MN) downstream of the droplet growth region to assess the droplet growth efficiency. The CPC measured the number concentration of particles with a diameter of 2.5 nm to 3 µm, and the OPS measured the number concentration of particles within a diameter in the range of 300 nm to 10 µm. Measuring droplet size distribution and growth efficiency right at the exit of the condensational growth apparatus is impractical when using particle counters. To address this, the tubing used to transport the droplet stream from the droplet growth region to the particle counters was shielded with fiberglass woven tape to maintain the temperature of the droplet flow, preventing changes in droplet size distribution. Additionally, sharp bends and elbows were avoided to reduce wall losses and the tube length from the droplet growth region to the counters was identical to ensure similar inertial droplet losses for

both particle counters. Measured growth efficiencies would serve as conservative estimates in case of appreciable droplet wall loss. Along with the CPC and the OPS, an external vacuum pump was used downstream of the TCALC to control the inlet aerosol flowrate at 8, 9 or 10 L min⁻¹.

Within the TCALC, the collection of enlarged droplets relies on inertial impaction. Thus, droplet growth to $d_p > 1.4 \mu\text{m}$ is imperative. It is worth noting that optical particle counters often underestimate the size of water droplets (Hinds and Kraske 1986), and their true aerodynamic diameter is expected to be larger than their measured optical diameter (d_d). In this study, the measured droplet growth efficiency refers to particle activation and droplet growth to $d_d > 1.4 \mu\text{m}$. That corresponds to an estimated aerodynamic diameter $\geq 1.8 \mu\text{m}$ (Zervaki et al. 2024; Chien et al. 2016), sufficiently large for inertial collection. The droplet growth efficiency (η) was calculated as follows:

$$\eta = \frac{N_{OPS}}{N_{CPC}} \quad (1)$$

where N_{OPS} denotes the number concentration with $d_d > 1.4 \mu\text{m}$, as measured by the OPS, and N_{CPC} denotes the total number concentration as measured by the CPC.

1.2 Number concentration effect

The performance of the TCALC was also evaluated as a function of the particle number concentration of the inlet aerosol flow (Figure 3). Crystalline silica test aerosol was generated using a fluidized bed aerosol generator (model 3400A, TSI Inc., Shoreview MN). The generator used a fine, ground silica powder (Min-U-Sil@5; US Silica, Katy TX) as the source material. The size distribution of the generated silica particles at the outlet of the fluidized bed is shown in Figure 4. To control the number concentration of the aerosol, the generated aerosol flow was diluted with particle-free air downstream of the fluidized bed. The aerosol flowrate at the inlet of the collector

was fixed at 10 L min⁻¹, and the relative humidity (*RH*) and temperature of the aerosol stream (*T_a*) were approximately 41.5% and 22.5 °C, respectively. The temperature of the generated vapor (*T_{sat}*) and the growth tube wall (*T_{gt}*) was controlled at 85 °C and 0 °C, respectively.

A butanol-based Condensation Particle Counter (model CPC 3776, TSI Inc., Shoreview MN) was used downstream of the collector to quantify the fraction of the particles that were not captured by the TCALC. The CPC was also used upstream of the TCALC to measure the particle concentration at its inlet. The collection efficiency (*C.E.*) achieved through the TCALC was calculated as follows:

$$C.E. = \frac{N_i - N_o}{N_i} \quad (2)$$

where *N_i* is the number concentration of the aerosols measured at the inlet of the collector, and *N_o* is the number concentration of the aerosols that were not collected in the TCALC.

1.3 Comparison with direct filter collection

The collection efficiency of the TCALC was compared to that of the filter-based collection method. To conduct this comparison, we used the experimental setup shown in Figure 5. Crystalline silica was generated using a fluidized bed aerosol generator. The size distribution of the generated aerosol at the outlet of the fluidized bed is shown in Figure 4. The relative humidity (*RH*) of the aerosol stream ranged from 42.2% to 74.8%. The average temperature of the aerosol stream (*T_a*) was approximately 21.5 °C.

The crystalline silica was collected simultaneously by the TCALC at a flowrate of 10 L min⁻¹, and a reference 0.4-μm pore size, 37-mm polycarbonate filter (225-1609; SKC Inc., Eighty Four PA) at a flowrate of 10 L min⁻¹. The filter was placed in a closed-face filter cassette, collocated, and operated in parallel with the TCALC. A DustTrak™ DRX (model 8533, TSI Inc.,

Shoreview MN) was used in parallel with the TCALC and the filter cassette to monitor the mass concentration collected. When the collected particulate mass on the filter was estimated to have exceeded 100 µg, the reference filter was replaced during the collection process, to prevent sample “overloading” on the filter and a potential increase of the pressure drop (Raynor et al. 2011). The total mass collected on all the reference filters for each measurement was calculated by summing the individual filter gravimetric measurements.

Following the collection, both the droplet impactor and the collection vial of TCALC were removed to recover the collected crystalline silica particles. The droplet impactor was rinsed with 3 ml of isopropyl alcohol, allowing for the retrieval of trace particles adhering to the droplet impactor surface. The liquid rinse containing particles was combined with the collected suspension. The suspension was then vacuum-filtered through a 20 µm or 30 µm-pore-sized mesh nylon filter, with a diameter of 25 mm (NY2002500, NY3002500; Millipore Sigma, St. Louis MO) and a 0.4-µm pore size, 25-mm-diameter polycarbonate filter (225-1608; SKC Inc., Eighty Four PA). The mesh nylon filter ensured removal of any impurities collected, including traces from the conductive silicone rubber tubing used for the sampling. The polycarbonate filter was used to recover the silica particles. The collection vial was rinsed with an additional 3 ml of isopropyl alcohol and the rinse was also subjected to filtration. The 25-mm-diameter polycarbonate filter was then placed in a temperature and relative humidity-controlled environment along with the 37-mm-diameter reference filter used during the aerosol collection, to dry. A microbalance (XPR6U Microbalance; Mettler Toledo, Columbus OH) was used for weighing the filters. The analyte mass collected on the filters (m_p) was then calculated:

$$m_p = M_f - M_i \quad (3)$$

where M_f and M_i denote the final and initial filter mass, respectively.

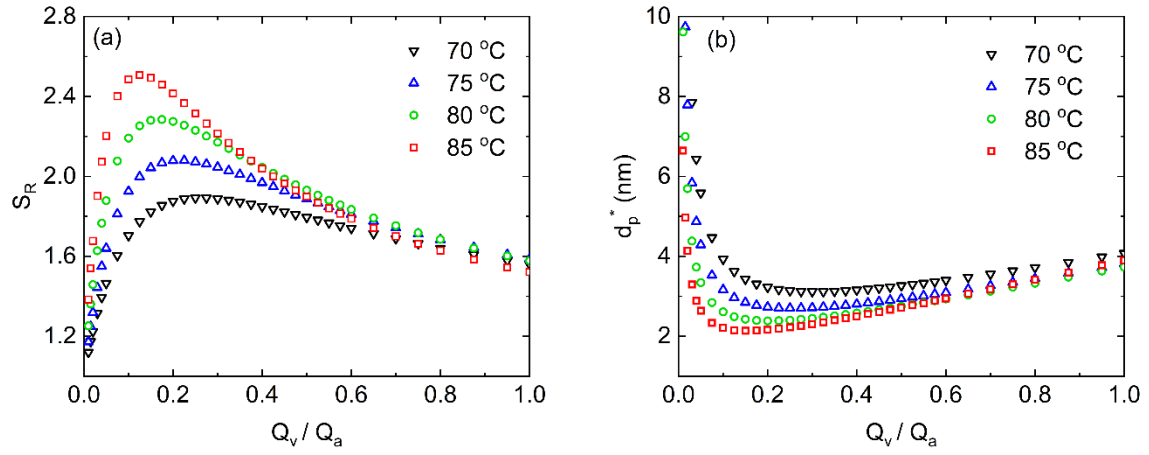


Figure 2 (a) Calculated saturation ratio (S_R) and (b) Kelvin diameter (d_p^*) as a function of the vapor-to-aerosol flowrate ratio, for a T_{sat} of 70, 75, 80, and 85 °C. The cold aerosol flow had a temperature of 22 °C, and 100 % RH was assumed.

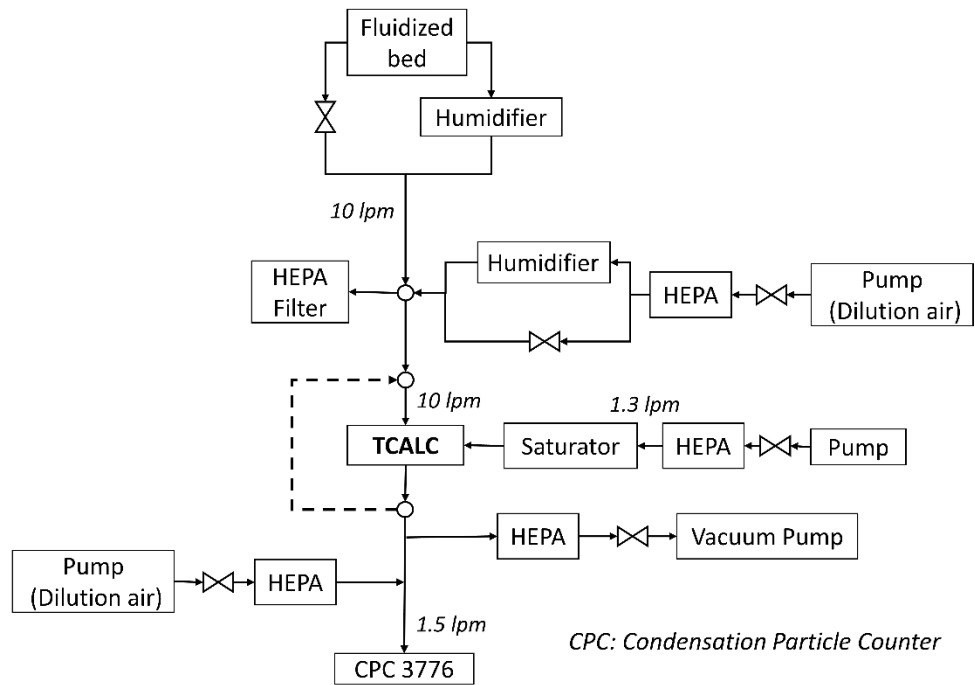


Figure 3 Experimental setup used for the generation of crystalline silica (Min-U-Sil@5) and the evaluation of the effect of number concentration on the TCALC.

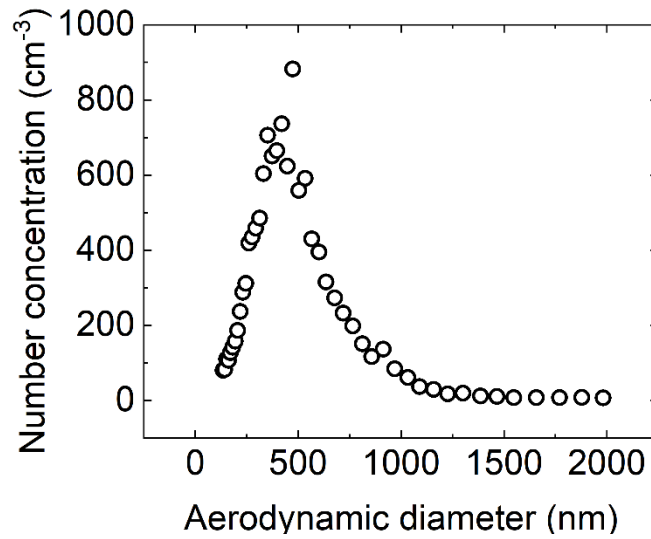


Figure 4 Size distribution of crystalline silica (Min-U-Sil@5) particles generated through the fluidized bed aerosol generator (TSI), as measured at the outlet of the aerosol generator using an Aerodynamic Aerosol Classifier (AAC, Cambustion) and a Condensation Particle Counter (CPC 3776, TSI) in series.

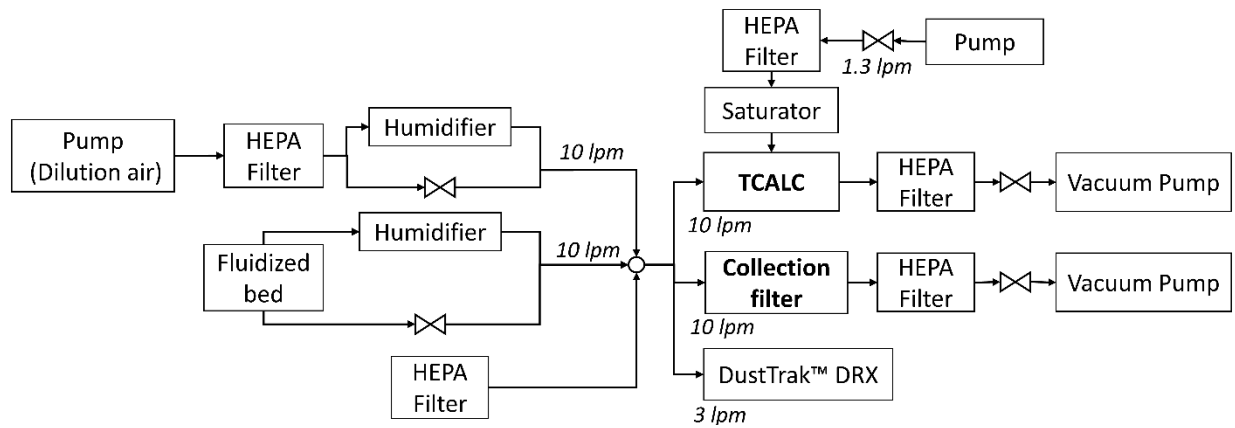


Figure 5 Experimental set up used for the generation of crystalline silica (Min-U-Sil@5) and the following collection using the TCALC and a reference 37-mm-diameter polycarbonate filter.