

1. Materials and Methods

1.1. Activation and growth efficiency

We measured the particle activation and droplet growth efficiency of the TCAC as a function of five parameters: (1) the mixing ratio (Q_h/Q_c), (2) the saturator temperature (T_h), (3) the relative humidity of the aerosol flow, (4) the particle diameter of the sampled aerosol (d_p), and (5) the number concentration of the sampled aerosol. The dry spot diameter and particle radial distribution were also determined by collecting particles on a substrate downstream of the concentrator. The optimum window of operating parameters was then identified.

Figure 1 shows the experimental setup used for characterizing the TCAC. A small-volume medical nebulizer (Salter 8900 Series; Salter Labs, Arvin CA, USA) along with a diffusion dryer was used to generate sodium chloride test aerosol, at a flow rate of 3 – 4 L min⁻¹. An aerosol flow at atmospheric pressure was then introduced into the Aerodynamic Aerosol Classifier (AAC; Cambustion Ltd, Cambridge, United Kingdom) at a flow rate of 1.5 L min⁻¹, to generate a near-monodisperse test aerosol. Particle-free, dilution air was added downstream of the AAC to increase the total aerosol flow rate to 4 L min⁻¹, which was introduced into the TCAC. The humidity of the aerosol flow was varied using commercial NafionTM humidifiers (model MH-110-12F-4; Perma Pure LLC, Lakewood NJ, USA) to control the relative humidity (RH) at 11, 15, 25 or 100 %. No other RH conditions between 25% and 100% were explored due to similar growth efficiencies obtained under both conditions for saturator temperatures ≥ 75 °C. Particle-free flow was also introduced into the saturator of the TCAC to generate hot saturated flow. The saturator temperature was set to 70, 75, 80 or 85 °C, and the flow rate was set to 0.2, 0.4, 0.6, 0.8, 1.0, 1.2, 1.4 or 1.6 L min⁻¹. The flow rate of the hot saturated flow was measured at the inlet of the saturator at 22 °C. The flow rate across the TCAC was controlled within the range of 4.2 – 5.6 L min⁻¹ by a vacuum pump and two particle counters: the Ultrafine Water-based Condensation Particle Counter (model 3786 UCPC; TSI Inc., Shoreview MN, USA) and the Optical Particle Sizer (model 3330 OPS; TSI Inc., Shoreview MN, USA).

The particle/droplet number concentration downstream of the TCAC was measured using the two particle counters. The UCPC provided a total number concentration of particles with a diameter in the range of 2.5 nm to $> 3 \mu\text{m}$, while the OPS classified particles into 16 different channels in the size range of 300 nm to $10 \mu\text{m}$ based on their optical diameter. The OPS was connected in parallel with the UCPC to detect and count the number of grown droplets and the final droplet optical diameter (d_d). The tubing transporting the aerosol from the TCAC outlet to the particle counters was thermally shielded using fiberglass woven tape to ensure that there was no change in the droplet size distribution. Moreover, the residence time of the particles/droplets in the TCAC growth tube was estimated to be $> 0.1 \text{ s}$, providing sufficient time for activation and growth of all particles in the sample flow. Therefore, the droplets grew to collectable sizes before exiting the growth tube of the TCAC. The length of tubing from the outlet of the TCAC to the inlet of OPS and UCPC were kept identical to ensure similar transport losses.

For this study, we calculated the minimum collectable aerodynamic diameter (at which droplets could be collected by impaction), which was approximately $\geq 1.4 \mu\text{m}$ (d_p). Optical particle counters are typically calibrated with PSL spheres, which have a refractive index of 1.6. A pure water droplet has a refractive index of 1.33. As a result, water droplets are expected to appear undersized when detected by optical counters (Garvey and Pinnick 1983; Hinds and Kraske 1986; Pinnick, Garvey, and Duncan 1981). In this study, NaCl particles with an aerodynamic diameter (d_p) of $\geq 25 \text{ nm}$, were enlarged into droplets with $d_d > 1400 \text{ nm}$. Assuming that the droplets were spherical, and made of pure water, the aerodynamic diameter was estimated to be $1.8 \mu\text{m}$ from Chien et al. (2016). However, the equation provided by Chien et al. (2016) was developed for oleic acid aerosols, which have a refractive index of 1.46, lower than that of PSL but larger than that of water. Therefore, the actual aerodynamic diameter of the water droplets is expected to be larger than $1.8 \mu\text{m}$. As a result, the growth efficiency reported in this study, which accounted for both the nuclei activation and the droplet growth efficiency, was based on optical diameters greater than $1.4 \mu\text{m}$ (unless stated otherwise), and was expressed as follows:

$$\eta = \frac{N_{1.4}}{N_T} \quad (1)$$

where η denotes the calculated growth efficiency, $N_{1.4}$ denotes the number concentration of droplets with $d_d > 1.4 \mu\text{m}$ measured by the OPS, and N_T denotes the total number concentration of particles and/or droplets measured by the UCPC.

1.2. Wall losses

Wall losses within the TCAC were evaluated, in the absence of particle growth, by measuring the particulate mass, determined using gravimetric measurements, of the aerosol entering and exiting TCAC. Min-U-Sil@5 (US Silica, Katy TX, USA) and Min-U-Sil@10 (IIT Research Institute, Chicago IL, USA) were used as test particles. The aerosol sample flow was maintained at a flow rate of 4 L min^{-1} through the TCAC, while the saturated flow, entering from the side of the mixing “tee”, had a flow rate of 0.6 L min^{-1} , all at room temperature to avoid particle growth. Downstream of the concentrator, a filter was positioned to capture the spot sample from the mixed flow exiting the TCAC and was subsequently used to determine the collected particulate mass. A parallel filter collection was performed to assess the particulate mass concentration of the aerosol entering the TCAC at identical conditions. The difference between the two particulate mass measurements was taken as the total wall loss within the TCAC system. No heating was applied at the saturator to prevent filter saturation and minimize pressure drop.

1.3. Spot deposit diameter

To measure the deposit diameter obtained using the TCAC, polystyrene nanospheres of 20 nm, 150 nm diameter (NIST Traceable Size Standards, Thermo Fisher Scientific, Waltham MA, USA) and 1.9- μm -diameter fluorescent beads (fluoro-max green beads; Thermo Fisher Scientific, Waltham MA, USA) were used as seed particles. The nano and microsphere aerosols were generated from liquid suspensions. The particles encapsulated within the grown droplets exiting the TCAC were collected on a flat heated surface at $90 - 100 \text{ }^\circ\text{C}$ ($T_{\text{substrate}}$) downstream of the TCAC, located at the experimentally determined optimum nozzle-to-plate distance of 4 mm. The droplets impacted directly onto an aluminum-backed, carbon tape

(product 16086-5; Ted Pella Inc., Redding CA, USA) that could be readily analyzed using Scanning Electron Microscopy (SEM; Phenom XL Desktop SEM, Thermo Fisher Scientific, Waltham MA, USA) to obtain the dry spot diameter and the spatial distribution characteristics within the spot. The dry spot diameter, defined as the diameter of the circle encompassing 90 % of the deposited particles, was calculated by determining the radial distribution of the projected area of the collected particles on the substrate, using the ImageJ software (Schneider, Rasband, and Eliceiri 2012). We expect negligible thermophoretic losses during droplet impaction, primarily due to the high velocity of the aerosol jet (> 30 m/s) and the very small thermal gradient to induce appreciable thermophoresis.

1.4. Number concentration effect.

The number concentration of the test aerosol at the inlet of the concentrator was varied to investigate its impact on the performance of the TCAC. Dilution flow or make-up air was used to achieve the desired number concentrations. To minimize any effects on the performance of the concentrator due to the dilution, the mixing ratio was kept constant across the range of number concentrations tested.

1.5. Counting Statistics for Fiber Concentration Measurement

The application of TCAC to the measurement of airborne fiber concentrations was investigated. Fiber counting using Phase Contrast Microscopy (PCM) is a common method for quantifying fiber concentration collected on a filter. The NIOSH method 7400 recommends collecting fibers on 25-mm filters for PCM analysis (NIOSH 2019); however, other collection techniques can also be used to concentrate the fiber sample in a small spot deposit, reducing counting uncertainty and sampling time. Shorter collection times are essential for rapid detection of airborne fibers, such as asbestos fibers, given their potential hazard even at low concentrations (NIOSH 2019). We estimated the sampling time required to achieve a targeted Poisson counting uncertainty for fiber concentration measurements using PCM for different collection methods: TCAC, Sequential Spot Sampler, and filter-based collection.

Sampling of a large number of fibers, n , can reduce the counting uncertainty ($\sigma \% = 1/\sqrt{n}$). For PCM analysis, the NIOSH method 7400 recommends a substrate area of 0.785 mm^2 (A_m), and a fiber density of $100\text{--}1300 \text{ mm}^{-2}$, for optimal and unbiased counting ($\sigma = 2.8 - 10 \%$; NIOSH 2019). Assuming a fiber concentration of 0.1 cm^{-3} (c_f), the required sampling time (t_c) to achieve a target counting uncertainty of $3 - 10 \%$ was calculated:

$$t_c = \frac{n A_d}{A_m c_f Q \eta_c} \quad (2)$$

where A_d is the spot deposit area of each collection technique, Q is the sample flow rate employed in each collection method, and η_c is the collection efficiency. The operating parameters of the aerosol collection methods used for the calculation of the required sampling time are listed in Table 1. The spot sample generated from TCAC has an area of approximately 1.54 mm^2 , while the effective collection area of the Sequential Spot Sampler and the 25-mm filter are 0.785 and 385 mm^2 , respectively.

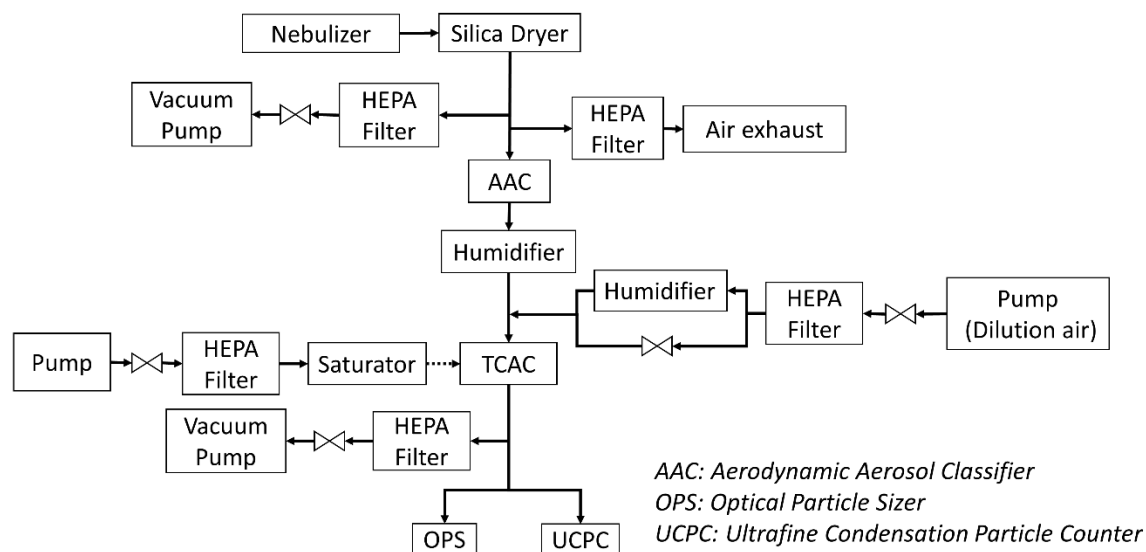


Figure 1. Experimental set up used for the growth efficiency measurements. Solid line arrow denotes flow of air stream. Dashed line arrow denotes flow of hot vapor-saturated stream.

Table 1. Operating parameters of aerosol collection techniques for calculation of the Poisson counting statistics of fiber concentration measurement using the phase contrast microscopy (PCM).

<i>Collection method</i>	Collection Efficiency	Sample Flow Rate (L min⁻¹)	Spot sample diameter (mm)
<i>TCAC</i>	0.86	4	1.4
<i>Sequential Spot Sampler^{TM, 1}</i>	0.95	1.5	1.0
<i>25-mm-filter</i>	1.0	2	22

¹Performance parameters obtained from manufacturer.

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