

A high-throughput, Turbulent-mixing, Condensation Aerosol Concentrator for Direct Aerosol Collection as a Liquid Suspension

Introductory Information

Trace measurement of aerosol chemical composition in workplace atmospheres requires the development of high-throughput aerosol collectors that are compact, hand-portable, and can be operated using personal pumps. We describe the design and characterization of a compact, high flow, Turbulent-mixing Condensation Aerosol-in-Liquid Concentrator (TCALC) that allows direct collection of aerosols as liquid suspensions, for off-line chemical, biological, or microscopy analysis. The TCALC unit, measuring approximately $12 \times 16 \times 18$ cm, operates at an aerosol sample flowrate of up to 10 L min^{-1} , using rapid mixing of a hot flow saturated with water vapor and a cold aerosol sample flow, thereby promoting condensational growth of aerosol particles. We investigated the effect of operating parameters such as vapor temperature, growth tube wall temperature, and aerosol sample flowrate, along with the effect of particle diameter, inlet humidity, aerosol concentration, and operation time on TCALC performance. Nanoparticles with an initial aerodynamic diameter ≥ 25 nm could grow to droplet diameters > 1400 nm with an efficiency $\geq 80\%$. Good droplet growth efficiency was achieved for sampled aerosol relative humidity $\geq 9\%$. We measured complete aerosol collection for concentrations of $\leq 3 \times 10^5 \text{ cm}^{-3}$. The results showed good agreement between the particulate mass collected through the liquid collector and direct filter collection. The TCALC eliminates the need for sample preparation and filter digestion during chemical analysis, thereby increasing sample recovery and substantially improving the limit of detection and sensitivity of off-line trace analysis of collected liquid samples.

Experimental Methods

Experiments were designed and conducted for the evaluation of the TCALC based on the following key variables:

- (i) The temperature of the vapor flow generated through the saturator (T_{sat}),
- (ii) The temperature of the growth tube wall (T_{gt}).
- (iii) The aerodynamic diameter (d_p) of the sampled aerosol, and
- (iv) The relative humidity of the aerosol flow.

The performance of the TCALC was also evaluated as a function of the particle number concentration of the inlet aerosol flow. Finally, the collection efficiency of the TCALC was compared to that of the filter-based collection method.

Droplet Growth Efficiency

Particle generation:

Particles used for the measurement of the droplet growth efficiency were generated through a jet *nebulizer* (Salter 8900 Series Disposable Small Volume Jet Nebulizer; Salter Labs, Arvin CA):

- Liquid solutions of sodium chloride were used.
- A diffusion dryer (model 3062, TSI Inc., Shoreview MN) containing silica gel desiccant, was used downstream of the nebulizer for removal of water vapor and reduction of the dew point of the aerosol stream.

Particle classification:

- The Aerodynamic Aerosol Classifier (AAC; Cambustion Ltd, Cambridge, United Kingdom) was used to achieve near-monodisperse (in aerodynamic size) test aerosol with $d_p = 25, 50, 75, 100, 150, 200, 250$ and 300 nm, at a controlled flow rate of 1.5 L min^{-1} .

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Efficiency measurements:

- The TCALC was evaluated for aerosol flow rates of 8, 9 and 10 L min⁻¹. To attain these flow rates, we used a valve-controlled, particle-free dilution air flow downstream of the AAC.
- Upstream of the TCALC, two humidifiers (model 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 collector at an ambient temperature of 22 °C, and an RH in the range of 9 – 60 %.
- Particle-free air was introduced into the saturator, and a vapor-saturated flow was produced at a controlled flow rate of 1, 1.2, 1.3, 1.4, 1.6, 1.8, 2, 2.3 or 2.5 L min⁻¹. The saturator temperature was set at a fixed temperature of 70, 75, 80 and 85 °C.
- 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.
- The OPS measured the number concentration of particles within a diameter in the range of 300 nm to 10 µm.

- 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.
- 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.

Droplet Optical Diameter (d_d) and Aerodynamic Diameter (d_p):

- 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.
- 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.

Number concentration effect.

Particle generation:

Particles used for the evaluation of the particle number concentration effect on the collector, were 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 to generate respirable crystalline silica.
- To control the number concentration of the aerosol, the generated aerosol flow was diluted with particle-free air downstream of the fluidized bed.

Efficiency measurements:

- The aerosol flow rate 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.

Comparison with direct filter collection

Particle generation:

Particles used for the comparison with direct filter collection, were generated using a *fluidized bed aerosol generator* (model 3400A, TSI Inc., Shoreview MN):

- The generator used a fine, ground silica powder (Min-U-Sil®; US Silica, Katy TX) as the source material to generate respirable crystalline silica.
- 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.

Sample collection and retrieval.

- 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.

Citations- Publications based on the dataset.

Zervaki, O., Dionysiou, D. D., & Kulkarni, P. (2024). A high-throughput, turbulent-mixing, condensation aerosol concentrator for direct aerosol collection as a liquid suspension. *Journal of Aerosol Science*, 182, 106442. <https://doi.org/10.1016/j.jaerosci.2024.106442>

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