



Wet Cyclone Sampling for Time-Resolved Molecular Analysis of Organic Aerosols: Performance Evaluation and Application to Incense Smoke

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1 Abstract

2 This study was motivated by the need for a practical alternative to conventional filter sampling
3 that retains its ease of field deployment while providing improved temporal resolution. A
4 condensational growth wet cyclone sampler was developed and characterized for time-resolved
5 collection of organic aerosols and coupled with liquid chromatography mass spectrometry for
6 molecular analysis of aerosol composition. The sampler combines the condensational growth
7 chamber of a particle-into-liquid sampler with a miniature wet cyclone, where the grown droplets
8 are collected by centrifugal deposition into a continuously wetted liquid vortex. Collection
9 performance of the sampler was optimized using polydisperse oleic acid particles as a
10 representative of water-insoluble organic aerosols. Under optimized conditions, the collection
11 efficiency of oleic acid particles reached $64.5 \pm 8.3\%$, with remaining losses attributed primarily to
12 liquid entrainment into the gas exhaust and compound-specific retention within the collection
13 pathway. Collection efficiency was relatively insensitive to the initial particle size distribution over
14 the range examined, as condensational growth produced droplets sufficiently large for effective
15 cyclone collection. The sampler was subsequently applied to incense smoke, where its recovery
16 of molecular components was compared with conventional filter sampling under the same mass-
17 spectrometric analysis. The cyclone samples captured major molecular features observed in paired
18 filter samples and also recovered molecules extending to relatively high carbon numbers.
19 Sequential collection further resolved the temporal evolution of particulate diethyl phthalate, a
20 major component of incense smoke. These results demonstrate that the condensational growth wet
21 cyclone broadens liquid-based collection to a wider range of particle-phase constituents and
22 provides an accessible interface for time-resolved molecular characterization of organic aerosols.

23 1. Introduction

24 The environmental and health impacts of organic aerosol (OA), a substantial and ubiquitous
25 fraction of atmospheric particulate matter, are closely linked to its molecular composition (Pöschl,
26 2005; Shiraiwa et al., 2017; Pye et al., 2021). OA comprises hundreds to thousands of organic
27 compounds originating from both direct emissions and atmospheric transformation of volatile
28 organic precursors (Hallquist et al., 2009; Jimenez et al., 2009). These compounds span broad
29 ranges of molecular weight, functionality, solubility, and volatility, and are heterogeneously
30 distributed among individual particles or internally mixed with inorganic constituents. The OA
31 composition varies substantially among natural and anthropogenic sources and continues to evolve
32 after emission through photochemical processing, gas-particle partitioning, and condensed-phase
33 chemistry (Pöschl and Shiraiwa, 2015). For combustion-derived OA, this variability is further
34 affected by fuel composition, combustion efficiency, fire intensity, and subsequent atmospheric
35 aging (Reid et al., 2005). The molecular composition of OA governs key physicochemical
36 properties, including volatility, hygroscopicity, phase state, optical behavior, and chemical
37 reactivity (Koop et al., 2011; Moise et al., 2015; Zhang et al., 2015a; Reid et al., 2018). These
38 properties, in turn, influence its atmospheric lifetime, radiative effects, cloud-forming potential,
39 and toxicity (Mauderly and Chow, 2008; Goldstein et al., 2009; Shrivastava et al., 2017).
40 Molecular-level characterization of OA is therefore essential for resolving its chemical diversity,
41 linking composition to physicochemical properties, and assessing its climate and health effects.

42 Recent advances in online mass spectrometry have enabled molecularly resolved
43 measurements of OA at temporal resolutions on the order of seconds to an hour. These approaches



44 can be broadly classified as collection-based methods and direct-inlet methods (Nizkorodov et al.,
45 2011; Laskin et al., 2012; Noziere et al., 2015; Johnston and Kerecman, 2019; Zhang et al., 2024;
46 Sun et al., 2025). Collection-based methods, including filter inlet for gases and aerosols chemical
47 ionization mass spectrometry (FIGAERO-CIMS) (Lopez-Hilfiker et al., 2014; Chen et al., 2020;
48 Thornton et al., 2020), micro-orifice volatilization impactor chemical ionization mass
49 spectrometry (MOVI-CIMS) (Yatavelli and Thornton, 2010; Yatavelli et al., 2012; Mohr et al.,
50 2013), and thermal desorption aerosol gas chromatography mass spectrometry (TAG-GC/MS)
51 (Williams et al., 2006; Kreisberg et al., 2009; Williams et al., 2010; Kristensen et al., 2023), first
52 collect and concentrate aerosol particles on an impaction surface over a defined sampling interval.
53 The deposited material is then thermally desorbed under a controlled carrier-gas flow and analyzed
54 either following chromatographic separation, as in TAG-GC/MS, or directly by chemical
55 ionization mass spectrometry, as in MOVI-CIMS and FIGAERO-CIMS. These methods provide
56 compound-specific information and, through temperature-programmed desorption in MOVI and
57 FIGAERO, can also characterize the volatility and thermal behavior of detected compounds. The
58 measured molecular profiles may be influenced by surface interactions, gas-phase adsorption, and
59 thermal decomposition (Williams et al., 2016; Stark et al., 2017). The temporal resolution typically
60 ranges from tens of minutes to one hour because each measurement requires a discrete particle-
61 collection period followed by thermal desorption and mass-spectrometric analysis. Direct-inlet
62 methods introduce aerosol particles continuously without a discrete collection step, thereby
63 enabling measurements at second-level time resolution. In the chemical analysis of aerosol online
64 coupled with proton-transfer-reaction mass spectrometry (CHARON-PTR-MS) (Eichler et al.,
65 2015; Muller et al., 2017; Gkatzelis et al., 2018) and the vaporization inlet for aerosols coupled
66 with nitrate chemical ionization mass spectrometry (VIA-NO₃-CIMS) (Häkkinen et al., 2023;
67 Zhao et al., 2023; Zhao et al., 2024), particles are transferred into a heated inlet, rapidly vaporized,
68 and subsequently detected by gas-phase ionization mass spectrometry. Extractive electrospray
69 ionization mass spectrometry (EESI-MS) uses a different approach, in which particle-phase
70 compounds are extracted into charged solvent droplets and softly ionized with limited thermal
71 decomposition and fragmentation (Gallimore et al., 2017; Lopez-Hilfiker et al., 2019; Pospisilova
72 et al., 2020). These methods can capture rapid variations in particle composition through
73 molecular-level characterization of individual OA constituents at high temporal resolution. The
74 breadth and quantitative representation of the detected compounds may be influenced by particle-
75 transmission efficiency, thermal decomposition in heated inlets, and compound-dependent
76 extraction and ionization responses.

77 Traditional filter-based sampling followed by offline liquid chromatography mass
78 spectrometry (LC/MS) remains widely used for molecular characterization of OA, despite being
79 more labor-intensive and time-consuming than online spectrometric methods. Filter samplers
80 are compact and readily deployable in field settings, and the broad availability of LC/MS platforms
81 supports their continued use in laboratory and field studies. A major limitation, however, is the
82 lack of temporal resolution associated with time-integrated sample collection and subsequent
83 offline analysis. This limitation has motivated the development and evaluation of intermediate
84 approaches that retain the operational simplicity of filter sampling while providing greater
85 temporal resolution than conventional offline methods. Our previous work coupled a particle-into-
86 liquid sampler (PILS) with LC/MS for time-resolved molecular characterization of chamber-
87 derived OA (Zhang et al., 2015b; Zhang et al., 2016; Zhang et al., 2017; Hao et al., 2025). Its
88 applicability is largely restricted to highly oxygenated OA, as collection efficiencies are very low
89 for low-polarity and water-insoluble particles. In the present study, the condensational growth



chamber of the PILS was coupled with a miniature wet cyclone to extend OA collection across a broader range of water solubilities. Within the condensational growth chamber, aerosol particles rapidly grow into micrometer-sized droplets through water vapor condensation under supersaturated conditions (Weber et al., 2001; Sorooshian et al., 2006). The resulting droplets subsequently enter the miniature wet cyclone (Orsini et al., 2008), where the combined action of rotating airflow and washing solution maintains a liquid vortex along the inner collection surface and continuously transfers the captured OA material into sequential vials for time-resolved analysis. This configuration was designed to enhance collection performance through centrifugal deposition and retention of droplets on a wetted cyclone wall. While a recent study has demonstrated the direct coupling of a wet cyclone with mass spectrometry for real-time characterization of water-soluble OA (Lau et al., 2025), the present study focuses on the collection performance of OA constituents spanning a broad range of solubility and polarity. The condensational growth wet cyclone was first characterized and optimized using oleic acid and *cis*-pinonic acid aerosols as model compounds with contrasting physicochemical properties. The optimized sampler was then applied to incense smoke, with the molecular composition of cyclone-collected samples benchmarked against a paired filter sample analyzed under identical LC/MS conditions. Sequential cyclone samples were further used to characterize the temporal profile of diethyl phthalate (DEP), a major component of incense smoke, to demonstrate the capability of this approach for time-resolved molecular measurements.

2. Experimental

2.1 Generation of pure-compound and incense-burning aerosols

Oleic acid (>99%, MilliporeSigma) and *cis*-pinonic acid (>98%, MilliporeSigma) were used as model compounds spanning low to moderate water solubility. Separate methanolic solutions of oleic acid and *cis*-pinonic acid, ranging from 0.01 to 0.1 M, were prepared and atomized in independent experiments to generate aerosols with different particle size distributions. Following desolvation, the resulting polydisperse aerosols spanning ~30–1000 nm were introduced into the 10 m³ Teflon chamber at the National Center for Atmospheric Research (NSF NCAR) (Zhang et al., 2018; Zhang et al., 2022; Moo et al., 2024). The chamber was maintained at a relative humidity below 5% and a temperature of ~22 °C. The chamber contents were mixed using pulsed air for ~30–40 min until the measured aerosol concentration stabilized. Particle size distributions and number concentrations were measured using a custom-built scanning mobility particle sizer consisting of a differential mobility analyzer (Model 3081, TSI Inc.) coupled to a condensation particle counter (Model 3010, TSI Inc.). Particle mass concentrations were calculated from the measured number size distributions by assuming spherical particles and applying the density of the corresponding pure compound. These calculated values were used as the reference concentrations entering the sampler for determination of the wet cyclone collection efficiency.

Incense-burning experiments were conducted at room temperature under dry and dark conditions in the 3 m³ stainless steel chamber at UC Merced (Moo et al., 2025). Before each experiment, the chamber was flushed with dry zero air generated by a precision zero-air generator (Peak Scientific) for ~24 h. Four incense sticks with different fragrances, purchased from Amazon (customer rating above 4.0 and more than 6,000 reviews), were burned simultaneously at the center of the chamber floor. Particle mass concentrations generated by incense burning were monitored using an Air Quality Egg® equipped with a Plantower PMS5003 laser sensor. Incense smoke was sampled through two parallel flows drawn at the same flow rate from a common chamber outlet



134 and directed to the wet cyclone and a quartz membrane filter (47 mm, Millipore), respectively.
135 Samples collected by the wet cyclone were analyzed directly by LC/MS without further
136 preparation. Compounds collected on the quartz membrane filter were extracted with isopropanol,
137 and the extracts were concentrated under a gentle stream of nitrogen to a final volume of 0.5 mL
138 before LC/MS analysis.

139 **2.2 Condensational growth wet cyclone sampler**

140 The condensational growth wet cyclone sampler was adapted from a conventional Particle-
141 Into-Liquid Sampler (PILS) (Weber et al., 2001; Sorooshian et al., 2006; Zhang et al., 2015b;
142 Zhang et al., 2016), with the original impaction collector replaced by a wet cyclone while retaining
143 the condensational growth chamber (Orsini et al., 2008). A schematic of the system configuration
144 and sampling flow path is shown in Figure S1 in the Supplement. Sampled aerosol first passed
145 through a 1 μm cut-size impactor and an organic carbon denuder to remove gas-phase organic
146 vapors. A steam flow generated at 100 °C was adiabatically mixed with the cooler sampled air in
147 a condensation chamber to create supersaturated conditions, under which particles grew into
148 droplets larger than $\sim 1 \mu\text{m}$. The resulting droplet-laden sampling flow entered tangentially through
149 an inlet located near the upper side of the cyclone and established a rotating gas flow within the
150 conical body. The washing liquid at a flow rate of 0.43 mL/min was introduced through a second
151 inlet positioned approximately 180° opposite the gas inlet. The rotating gas flow entrained the
152 washing liquid and distributed it along the inner wall, forming a standing liquid vortex that
153 provided a continuously wetted collection surface. The aerosol droplets were transported toward
154 the wetted wall by centrifugal motion, while the collected particulate materials were incorporated
155 into the washing liquid and carried toward the outlet at the base of the cyclone. The particle-
156 depleted gas formed an upward inner vortex and exited through the central outlet at the top. The
157 liquid input and withdrawal rates were balanced to maintain the standing vortex and minimize
158 sample loss through the gas exhaust. The collected liquid was dispensed into amber glass vials
159 mounted on a rotating carousel operating at 5 min intervals, thereby providing time-resolved
160 samples for molecular-composition analysis. Alternatively, the collected liquid could be
161 continuously delivered to an automated sample loop coupled to a two-dimensional switching LC
162 valve (Model G4236A, Agilent) for sequential LC/MS analysis, with a time resolution determined
163 by the analysis cycle (typically ~ 15 –20 min).

164 **2.3 Chemical analysis by liquid chromatography mass spectrometry**

165 A liquid chromatography quadrupole time-of-flight mass spectrometer (LC/MS, Model G6530,
166 Agilent) equipped with an electrospray ionization (ESI) source was used to measure the pure-
167 component organic aerosols and conduct untargeted profiling of particulate emissions from
168 incense burning. Chromatographic separation was performed using a ZORBAX StableBond C18
169 reversed-phase column (2.1 x 150 mm x 5 μm , Agilent). For positive-ion mode, the mobile-phase
170 program was set as follows: 0 min: 100% solvent A (0.1% formic acid in 99% water and 1%
171 acetonitrile) and 0% solvent B (0.1% formic acid in 90% acetonitrile and 10% water); 9–11 min:
172 0% solvent A and 100% solvent B; 11–13 min: 0% solvent A and 100% solvent B; and 13–15 min:
173 100% solvent A and 0% solvent B. For negative-ion mode, the mobile-phase program was set as
174 follows: 0 min: 100% solvent A and 0% solvent B; 5–13 min: 0% solvent A and 100% solvent B;
175 13–15 min: 100% solvent A and 0% solvent B. The MS conditions for both positive- and negative-
176 ion modes were as follows: 300 °C gas temperature, 12 L/min drying gas flow, 35 psi nebulizer
177 gas, 275 °C sheath gas temperature, 12 L/min sheath gas flow, 3500 V capillary voltage, 250 V
178 nozzle voltage, 100 V fragmentor voltage, and 65 V skimmer voltage. The mass-to-charge ratio



(m/z) scale was calibrated daily using the ESI-L tuning mix (G1969-85000, Agilent), and mass correction in individual samples was simultaneously performed using the reference mass standard kit (G1969-85003, Agilent). Molecular features in the incense-burning samples were extracted using Agilent MassHunter Qualitative Analysis software (version 10.0). The pure-component organic aerosols collected by the wet cyclone were quantified using external calibration curves generated from their respective authentic standards. Diethyl phthalate in the incense-smoke samples was quantified using an external calibration curve generated from an authentic standard.

3. Results and Discussions

3.1 Performance characterization of the condensational growth wet cyclone

3.1.1. Optimization of operating conditions and performance evaluation

The overall mass collection efficiency (η_i) of the condensational growth wet cyclone was defined as the fraction of aerosol mass entering the sampler that was recovered in the cyclone collection liquid. In this study, η_i was determined experimentally using pure-component aerosols generated by atomizing a methanolic solution of a given model compound i into the chamber. The aerosol mass concentration entering the sampler was calculated from the SMPS-measured particle number size distribution in the well-mixed chamber (dN_i/dD_p) _{j} , weighted by the volume of a spherical particle with diameter $D_{p,j}$ and the density of the compound (ρ_i), and integrated across the measured particle size range. The mass concentration of compound i in the cyclone collection liquid (C_i) was quantified by LC/MS using compound-specific external calibration.

$$\eta_i = \frac{Q_l C_i}{Q_g \sum_j \left(\frac{dN_i}{dD_p} \right)_j \rho_i \Delta D_{p,j} \frac{\pi}{6} D_{p,j}^3} \quad (1)$$

where Q_l is the washing liquid flow rate and Q_g is the gas sampling flow rate. The measured collection efficiency may deviate from unity because of incomplete condensational activation or droplet growth, loss of droplets or collection liquid with the outlet gas, and retention of collected material on internal surfaces. Accordingly, operating parameters including the gas sampling flow rate and washing liquid flow rate may affect collection efficiency by controlling condensational growth, cyclone capture, and liquid-vortex stability. Washing liquid composition may further influence collection performance through its effects on wall wetting and analyte recovery.

Figure 1 presents the optimization of the gas sampling flow rate and washing liquid composition for maximizing the collection efficiency of oleic acid, which was selected as a model compound representative of highly water-insoluble organic aerosols. Across gas sampling flow rates of ~9–20 L/min, collection efficiency peaked at 15.5 L/min, decreasing gradually by approximately one-third at 9.8 L/min but falling sharply to approximately one-third of the maximum as the flow rate increased to 19.8 L/min. This nonmonotonic response reflects a balance between the efficiency of condensational droplet growth and the strength of the gas-driven liquid vortex required for effective cyclone capture. At lower flow rates, the tangential gas velocity within the cyclone may have been insufficient to sustain a stable liquid vortex, limiting droplet deposition and retention. At higher flow rates, increased gas velocity enhanced the entrainment of washing liquid and collected droplets into the exhaust, as evidenced by the substantially lower liquid volume recovered under these conditions. A similar dependence on sampling flow rate was observed when water was used as the washing liquid, with collection efficiency likewise reaching a maximum at 15.5 L/min (Figure S2 in the Supplement). This consistency suggests that the



220 observed flow-rate dependence was primarily governed by cyclone hydrodynamics rather than by
221 isopropanol evaporation. Washing liquid composition had an even stronger influence, with the
222 highest collection efficiency observed using 100% isopropanol, an approximately 62% decrease
223 after the addition of 20% water, and near-zero recovery with 100% water. This trend is consistent
224 with the low water solubility of oleic acid, which is more effectively dissolved and transported
225 through the liquid pathway in isopropanol. Increasing the water fraction likely promoted the
226 retention of oleic acid as a surface film or separate hydrophobic phase within the cyclone and
227 transfer lines. These results therefore indicate that the measured collection efficiency for oleic acid,
228 as a representative water-insoluble aerosol, is governed not only by condensational growth and
229 cyclone capture but also by liquid-phase transfer and sample recovery. The washing liquid flow
230 rate was constrained by the operating settings of the steam-generation system. The peristaltic
231 multi-channel pump was first set to deliver the optimized steam generation water flow of 1.5
232 mL/min. At this fixed pump setting, the washing liquid flow rate was controlled by the tubing
233 diameter. The selected tubing provided a flow rate of 0.43 mL/min, which was sufficient to fully
234 wet the cyclone surface and maintain a stable liquid vortex, as shown in Video S1 in the
235 Supplement.

236 Under the optimized operating conditions, overall method performance was evaluated in terms
237 of analytical detection limit, repeatability, analyte recovery, carryover, and measurement
238 uncertainty. The analytical detection limit for oleic acid was 28.9 ng/mL in the collection liquid,
239 corresponding to an equivalent aerosol concentration of 0.41 $\mu\text{g}/\text{m}^3$ for a sampling duration of 3
240 min. Repeatability was assessed from three replicate experiments conducted under identical
241 conditions, yielding a relative standard deviation of 12.8% for the measured mass collection
242 efficiency. Analyte recovery was estimated to be 85.8% by introducing $\sim 210 \mu\text{M}$ oleic acid into
243 the isopropanol washing flow and quantifying the compound in the resulting collected liquid.
244 Carryover testing showed that residual oleic acid in the subsequent blank accounted for less than
245 7.0% of the preceding sample response, indicating measurable but limited retention within the
246 cyclone and transfer pathway. The 3D-printed polymeric construction of the cyclone may have
247 contributed to the observed carryover, as oleic acid could be retained on the plastic surface and
248 subsequently remobilized by fresh washing liquid and transferred into later samples. The overall
249 method uncertainty for the derived oleic acid aerosol concentration was estimated to be 13.5% by
250 propagating uncertainties in LC/MS quantification and sample liquid recovery. Variability in the
251 final collected liquid volume was the dominant source of uncertainty because of liquid entrainment
252 into the gas exhaust and losses during debubbling, even though the washing liquid flow entering
253 the cyclone was maintained at 0.43 L/min. It is important to note that the performance metrics
254 established for oleic acid may not be directly transferable to complex organic aerosol mixtures,
255 because differences among constituents in solubility and affinity for polymeric surfaces can
256 produce compound-dependent collection and recovery, as discussed in the following section.

257 3.1.2. Effects of aerosol composition and size distribution on collection efficiency

258 To evaluate the influence of aerosol water solubility on collection performance, methanolic
259 solutions of *cis*-pinonic acid and oleic acid were atomized in separate chamber experiments to
260 generate organic aerosols representing relatively water-soluble and water-insoluble compounds,
261 respectively. In the original PILS impactor configuration, collection efficiency approached unity
262 for *cis*-pinonic acid but was near zero for oleic acid across all washing liquid compositions tested.
263 Integration of the wet cyclone with the PILS condensational growth chamber reduced the
264 collection efficiency of *cis*-pinonic acid aerosol to $81.3 \pm 8.7\%$. This reduction was primarily



265 attributed to nonselective loss of washing liquid through entrainment in the gas outlet flow, as
 266 evidenced by the lower-than-expected collected liquid volume and the visible transport of liquid
 267 droplets into the exhaust line. Further evidence was provided by a recovery experiment in which
 268 a known concentration of *cis*-pinonic acid was introduced through the washing flow and its
 269 concentration remained essentially unchanged in the recovered liquid. In contrast, the original
 270 PILS impactor configuration recovered virtually no oleic acid aerosol, regardless of washing liquid
 271 composition. Integration of the wet cyclone increased its collection efficiency to approximately
 272 $64.5 \pm 8.3\%$ under optimized operating conditions. The remaining difference in collection
 273 efficiency between oleic acid and *cis*-pinonic acid was attributed to additional compound-specific
 274 losses. In the recovery experiment, only 85.8% of the oleic acid concentration introduced through
 275 the washing flow was recovered. This incomplete recovery may have resulted from retention of
 276 oleic acid on the 3D-printed polymeric cyclone surfaces and incomplete transfer into the collected
 277 liquid. Overall, comparison of the two model compounds indicated that the baseline collection
 278 efficiency of the wet cyclone system was approximately 80%, with the primary loss attributable to
 279 entrainment of liquid droplets into the gas exhaust line. Additional losses were compound
 280 dependent, being negligible for *cis*-pinonic acid but appreciable for the strongly hydrophobic oleic
 281 acid. Accordingly, under the conditions examined in this study, the $64.5 \pm 8.3\%$ efficiency
 282 measured for oleic acid provides a conservative lower-bound estimate for atmospheric aerosol
 283 mixtures that typically contain compounds spanning a range of water solubilities.

284 The influence of particles size distribution on collection efficiency was evaluated using *cis*-
 285 pinonic acid aerosols generated from atomizing methanolic solutions of different concentrations
 286 ranging from 0.01 to 0.1 M. Two polydisperse particle populations were examined: a bimodal
 287 distribution with peaks at ~ 120 and ~ 280 nm and a unimodal distribution with a peak at ~ 240 nm.
 288 No appreciable difference in collection efficiency was observed between the two distributions,
 289 indicating that collection performance was relatively insensitive to the particle size distribution
 290 over the range examined, as shown in Figure 2. This behavior is consistent with effective
 291 condensational growth, through which particles in both distributions grew into micrometer-sized
 292 droplets sufficiently large for efficient cyclone collection, as shown in Figure S3 in the Supplement.
 293 As a result, differences in the initial particle size distributions were largely diminished before the
 294 droplets entered the cyclone, leading to similar collection efficiencies across the two populations.
 295 Potential transport losses within the condensational growth chamber include diffusional deposition
 296 of the smallest particles and gravitational settling or inertial deposition of larger particles. Particles
 297 larger than approximately $1 \mu\text{m}$ were removed by the inlet impactor and therefore did not affect
 298 the comparison. Particles smaller than 30 nm were not quantified under the SMPS operating
 299 conditions used in this study. Nevertheless, their contribution to the calculated aerosol mass was
 300 expected to be minor because the mass distributions were dominated by particles with diameters
 301 of 100 nm or greater.

302 **3.2 Application of the condensational growth wet cyclone to incense smoke**

303 **3.2.1. Comparison of molecular composition with conventional filter sampling**

304 The molecular composition of incense aerosol collected by the wet cyclone was compared with
 305 that obtained using conventional filter sampling to evaluate how well direct aqueous-phase
 306 collection preserves aerosol molecular composition. For consistent comparison between the two
 307 methods, intensities of all molecular signatures identified by LC/MS were normalized to the total
 308 aerosol mass collected by each sample. As shown in Figure 3, both sampling approaches recovered
 309 a number of common molecular signatures spanning a broad range of carbon number (n_C) and



carbon oxidation state (OS_C). These shared compounds occupied identical positions in n_C - OS_C space, while differences in marker size and color reflect variations in LC/MS ion intensity between the two samples. These shared molecular features accounted for 73.8% and 48.0% of the total LC/MS ion intensity in the cyclone and filter samples, respectively, indicating that the two sampling approaches showed reasonable agreement in capturing the major molecular components of the incense aerosol. At the same time, the relative ion intensities of individual compounds were not identical between the two samples, which may reflect differences in collection efficiency and extraction recovery. For all shared molecular features, the ratio of normalized ion intensity in the wet-cyclone sample to that in the filter sample ranged from 0.46 to 1.80, with more than ~57% exhibiting ratios greater than unity. This distribution indicates a modest tendency toward higher relative recovery of individual molecular components by the wet cyclone compared with filter sampling. The comparison was restricted to molecular features detected by both methods with matching LC retention times, thereby minimizing differences associated with compound-specific ionization response and reducing potential ESI matrix effects.

Differences between the two sampling approaches were also observed for molecular features detected exclusively in either the cyclone or filter sample. As shown in Figure 3 and Tables S1 and S2 in the Supplement, the filter sample contained a greater number of molecular features than the wet-cyclone sample, with many occurring at relatively lower carbon numbers. This difference may partly reflect positive sampling artifacts associated with quartz-fiber filters, where semivolatile organic compounds can adsorb onto the filter substrate and subsequently be measured as particle-phase material. In addition, aerosols accumulated on the filter can provide an additional condensed-phase medium for absorptive partitioning of semivolatile compounds during sampling. The longer filter sampling period (15 min vs. 3 min for each cyclone sample) may further enhance accumulation of these species. Although LC/MS intensities were normalized to the total aerosol mass, this normalization does not account for compound-specific differences in collection and recovery between the two sampling methods. Conversely, the cyclone-specific features generally extended toward higher carbon numbers, including several compounds with n_C above 25. The carbon oxidation states of the method-specific compounds covered broadly similar ranges for the two sampling approaches. The recovery of relatively high- n_C compounds by the wet cyclone is noteworthy because such species generally have lower volatility and, depending on molecular functionality, may also exhibit lower water solubility. Their presence therefore indicates that direct aqueous-phase collection did not systematically exclude larger or less water-soluble organic molecules from the incense aerosol. The occurrence of these compounds exclusively in the cyclone sample may also reflect differences in post-collection recovery between the two approaches. Direct transfer of aerosol into the liquid phase avoids retention on the filter substrate and subsequent extraction steps, which may reduce losses of compounds that are strongly retained on quartz filters.

3.2.2. Temporal profile of diethyl phthalate in incense smoke

Diethyl phthalate (DEP) was the dominant molecular component detected in the cyclone-collected incense emissions, accounting for approximately 18.2% of the total ion intensity of all molecular features identified by LC/MS. DEP is a semivolatile phthalate ester that has been widely used as a plasticizer and a carrier for fragrance ingredients, and its occurrence in incense emissions has been reported previously (Tran and Marriott, 2007; Dalibalta et al., 2015; Song et al., 2023; Moo et al., 2025). The molecular identity of DEP collected by the wet cyclone was further confirmed by comparison of its characteristic mass-spectral behavior with that of an external DEP standard. Under positive electrospray ionization, DEP produced a prominent protonated molecular



ion ($[M+H]^+$) at m/z 223, together with a sodiated ion adduct ($[M+Na]^+$) at m/z 245. Subsequent collision-induced dissociation generated the characteristic fragment ion at m/z 149, corresponding to protonated phthalic anhydride, as shown in Figure 4A. In addition to molecular identification, sequential wet-cyclone samples were used to characterize changes in particulate DEP concentration over the course of incense burning. DEP concentrations increased approximately linearly during the initial 15 min of incense combustion, as shown in Figure 4B. A similar temporal pattern was observed for the concurrently measured $PM_{2.5}$ mass concentration. The close correspondence between these temporal profiles is consistent with DEP being a major component of the emitted particulate matter and indicates that its concentration changes tracked the buildup of fine-particle mass during combustion. These results further demonstrate that sequential wet-cyclone sampling can provide time-resolved molecular measurements of individual organic aerosol components and preserve the mass-spectral information required for compound identification.

4. Conclusions

This study was motivated by the need for time-resolved molecular measurements of organic aerosols in laboratory and field settings where online mass-spectrometric instrumentation may not be readily available. A condensational growth wet cyclone was therefore developed and evaluated as an alternative to conventional filter sampling. The sampler was adapted from a conventional particle-into-liquid sampler by replacing the impaction collector with a continuously wetted cyclone, thereby extending liquid-based collection to OA constituents spanning a broader range of water solubilities. The measured collection efficiency for oleic acid, used as a model compound representative of water-insoluble organic aerosol, was $64.5 \pm 8.3\%$ under optimized conditions. This overall efficiency can be attributed to a baseline collection efficiency of approximately 80%, primarily governed by liquid entrainment into the gas exhaust, combined with additional compound-specific effects associated with the incomplete recovery of the strongly hydrophobic oleic acid. Collection efficiency was relatively insensitive to the initial particle size distribution over the range examined, as condensational growth increased particles of different initial sizes to droplets sufficiently large for effective cyclone collection. Compared with the conventional PILS, which is primarily applicable to water-soluble OA, incorporation of the wet cyclone substantially improved the collection of water-insoluble OA and therefore broadened its applicability to more compositionally diverse atmospheric aerosols, including complex emissions such as biomass-burning OA. Application to incense smoke showed that the wet cyclone recovered major molecular components also observed by conventional filter sampling, with collected aerosol transferred directly into the liquid phase without requiring subsequent filter extraction. The recovery of relatively high-carbon-number compounds further indicated that liquid-based collection with the wet cyclone is not restricted to highly water-soluble OA constituents. Sequential collection additionally resolved the temporal evolution of particulate DEP during incense burning, supporting the application of the wet cyclone to time-resolved measurements of individual molecular components on a timescale of minutes. This configuration provides a flexible interface between aerosol collection and chemical analysis, with temporal resolution determined by the collection interval or downstream analytical cycle. The extension of liquid-based collection to OA constituents spanning a broader range of water solubilities supports time-resolved characterization of molecular composition across diverse OA systems.



398 **Supplement In formation.** Supplemental information to this article includes three figures, one
399 video, and two tables.

400 **Data Availability.** Data presented in this manuscript are available upon request to the
401 corresponding author.

402 **Author contributions.** Y. Wang: Methodology, Validation, Formal analysis, Investigation,
403 Visualization, Writing – original draft, Writing – Review & Editing. P. Hao: Validation, Formal
404 analysis, Investigation, Writing – original draft. Q. Ye: Validation, Formal analysis, Investigation,
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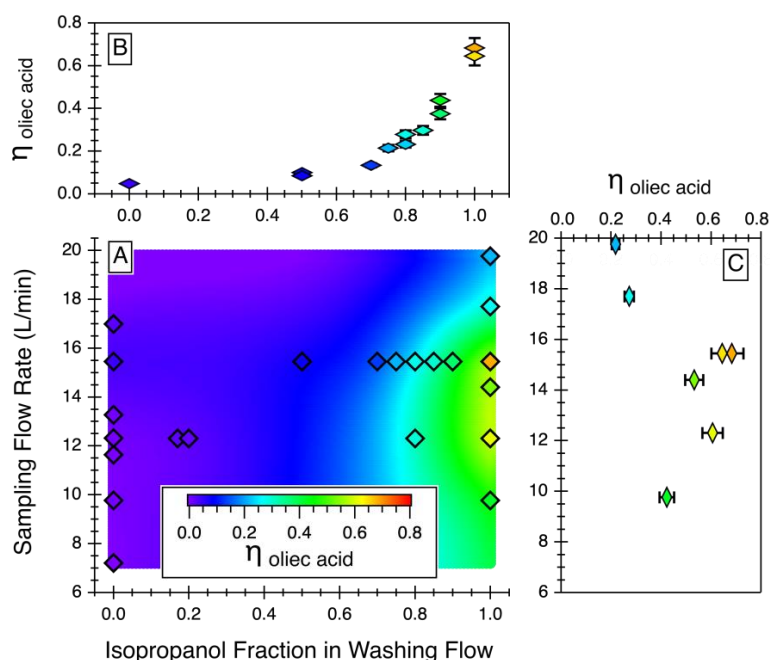


Figure 1. Optimization of wet-cyclone operating conditions for collection of oleic acid aerosol. (A) Oleic acid collection efficiency as a function of sampling gas flow rate and isopropanol fraction in the washing liquid. Diamond symbols represent measured collection efficiencies, and the background represents a smoothed interpolation of the measurements to illustrate the dependence of collection efficiency on the two operating parameters. (B) Oleic acid collection efficiency as a function of isopropanol fraction at a sampling gas flow rate of 15.5 L/min. (C) Oleic acid collection efficiency as a function of sampling gas flow rate with 100% isopropanol in the washing liquid.

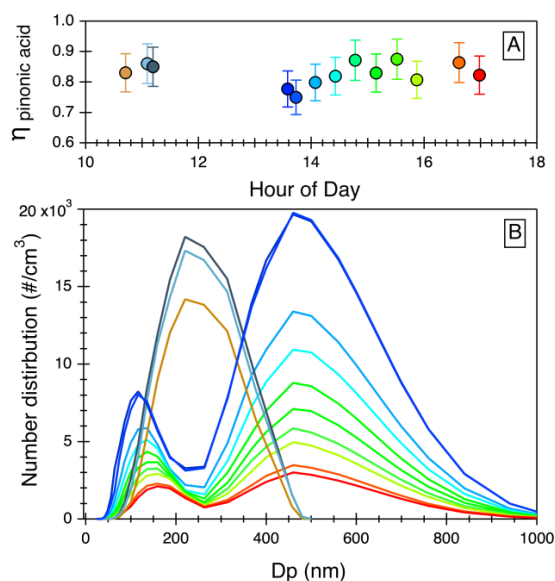


Figure 2. Effect of aerosol size distribution on the collection efficiency of *cis*-pinonic acid. (A) Collection efficiency measured at different times during two chamber experiments conducted on separate days. (B) Corresponding particle number size distributions measured by SMPS. Colors in panel B correspond to the collection-efficiency measurements in panel A at the same sampling times.

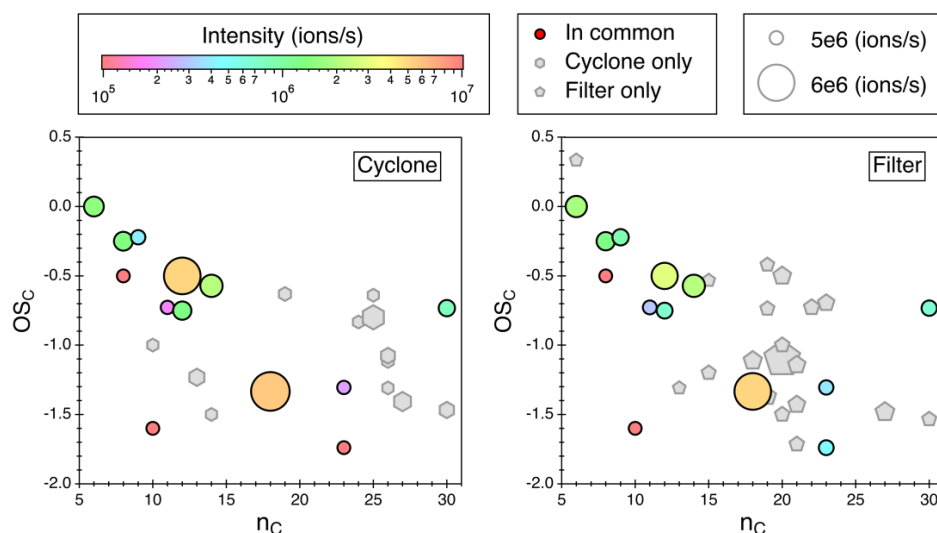


Figure 3. Molecular composition of incense smoke collected by the wet cyclone and conventional filter sampling. Detected molecular features are plotted according to carbon number (n_C) and oxidation state (OS_C) for (A) wet-cyclone and (B) filter samples. Colored circles denote molecular features detected by both sampling methods, whereas gray hexagons and pentagons denote features detected only in the cyclone and filter samples, respectively. Marker size represents LC/MS ion intensity for all detected molecular features, while marker color additionally indicates ion intensity on a logarithmic scale for features detected by both sampling methods.

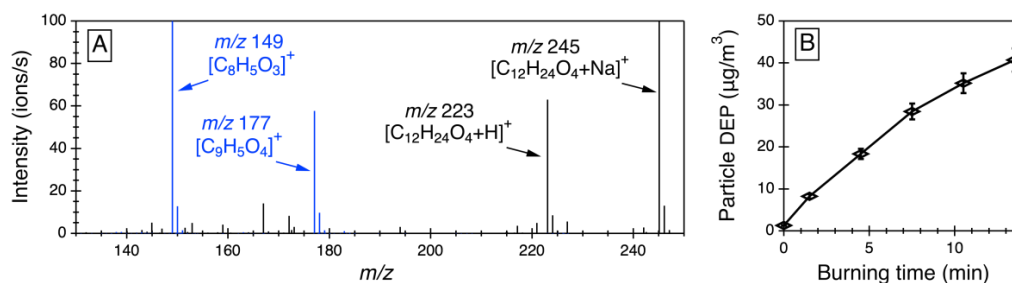


Figure 4. Identification and temporal evolution of particulate diethyl phthalate (DEP) during incense burning. (A) LC/MS spectrum of DEP, showing the protonated precursor ion at m/z 223 and the sodiated ion at m/z 245. The corresponding LC/MS/MS spectrum shows characteristic fragment ions at m/z 149 and m/z 177. (B) Time-resolved particulate DEP concentration during incense burning measured using sequential wet-cyclone collection.