

1 **Highly Viscous Phase Behavior of Organic-Rich Urban** 2 **PM_{2.5}**

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12 **Abstract**

13 Atmospheric aerosol viscosity strongly influences particle phase state, internal mixing, and multiphase chemical
14 processes, yet direct quantitative constraints for ambient urban PM_{2.5} remain limited. Here, we investigated the
15 phase behavior and viscosity of organic-rich PM_{2.5} samples collected during autumn 2023 from the urban
16 environments of Seoul and Beijing. Using filter extracts, relative humidity (RH)-dependent phase transitions and
17 morphological evolution of the droplets were examined by optical microscopy, revealing frequent two-phase and
18 three-phase morphologies during dehydration. Aerosol viscosity was quantitatively constrained at ~290 K under
19 experimentally accessible RH conditions (< ~45 %) using the poke-and-flow technique coupled with fluid-
20 dynamic simulations, yielding viscosities spanning from ~10⁴ to > ~10⁸ Pa·s. Compared with previously reported
21 laboratory-based viscosity measurements, the inferred viscosities of ambient PM_{2.5} were comparable to or
22 exceeded those reported for organic-rich ternary systems (i.e., sucrose–AS–H₂O and citric acid–AS–H₂O), which
23 are commonly used as laboratory proxy systems in aerosol viscosity studies. **These results indicate that organic-**
24 **rich urban PM_{2.5} can exhibit highly viscous, semisolid to solid phase states, and they provide quantitative, field-**
25 **based viscosity estimates constrained by the bulk organic and inorganic mass fractions of urban aerosols. Although**
26 **based on a limited number of filter samples and analyzed droplets, these findings offer a foundation for future,**
27 **more extensive viscosity studies of urban aerosols.**

28 Keyword: Viscosity, Morphology, PM_{2.5}, Organic Aerosol, Northeast Asia.

29 1. Introduction

30 Atmospheric fine particulate matter (PM_{2.5}) in urban megacities undergoes a range of general physicochemical
31 processes, including hygroscopic growth, phase transitions, viscosity changes, and diffusion-limited internal
32 mixing, which are strongly modulated by relative humidity (RH) (Swietlicki et al., 2008; Guo et al., 2014; Reid
33 et al., 2018; Song et al., 2022; Freedman et al., 2024; Tan et al., 2024). These processes play a critical role in gas–
34 particle partitioning (Zuend and Seinfeld, 2012; Zhou et al., 2013; Gkatzelis et al., 2018; Zaveri et al., 2020),
35 heterogeneous chemical reactivity (Li et al., 2020; Rasool et al., 2023; Song et al., 2025), aging dynamics
36 (Shiraiwa et al., 2011; Liu et al., 2025), and cloud formation (Suda et al., 2014; Cheung et al., 2020; Pöhlker et
37 al., 2023), yet their underlying mechanisms remain poorly constrained by empirical observations. This limitation
38 fundamentally undermines the accurate parametrization of urban aerosol impacts in atmospheric models, for
39 example, by violating the liquid-phase assumptions underlying gas–particle partitioning schemes and
40 heterogeneous reactive uptake coefficients (Shiraiwa and Seinfeld, 2012; Gržinić et al., 2015).

41 PM_{2.5} exerts major influences on air quality, human health, and climate forcing because of its chemical
42 complexity and diverse physical properties (Seinfeld et al., 2016; McNeill, 2017; Su et al., 2020; Nault et al., 2021;
43 Wall et al., 2022; El Haddad et al., 2024; Bei et al., 2025; Manavi et al., 2025; Sun et al., 2025). PM_{2.5} typically
44 consists of organic aerosols (OA), inorganic salts, trace metals, mineral dust, and elemental or black carbon (EC
45 or BC). Among these components, OA, including primary (POA) and secondary (SOA), have been frequently
46 observed as the dominant fraction across many continental regions, accounting for approximately 20–90 % of
47 total PM_{2.5} mass (Kanakidou et al., 2005; Hallquist et al., 2009; Jimenez et al., 2009; Huang et al., 2014; Zhang
48 et al., 2017; Jeon et al., 2023). These OA are chemically complex, encompassing thousands of molecules, and
49 their oxygen-to-carbon elemental ratios (O:C) have been shown to vary with atmospheric processing and
50 environmental conditions (Zhang et al., 2007; Aiken et al., 2008; Canagaratna et al., 2015; An et al., 2019;
51 Daellenbach et al., 2019; Sun et al., 2025; Cai et al., 2026).

52 Among the physical properties affected by the chemical complexity and atmospheric evolution of PM_{2.5},
53 **particle phase state, described in terms of dynamic** viscosity, is particularly important for controlling **particle**
54 reactivity. Viscosity **describes** the internal resistance of a material **to flow or deform**, quantifying molecular
55 mobility within the condensed phase. In the atmospheric context, viscosity serves as a critical indicator of particle
56 phase state: **aerosols with dynamic viscosities** below 10² Pa·s behave as liquids, those between 10² and 10¹² Pa·s
57 **are considered as amorphous semisolids**, and particles exceeding 10¹² Pa·s are **considered as amorphous** solids
58 (Koop et al., 2011; Zobrist et al., 2011). A highly viscous particle restricts internal diffusion and slows multiphase
59 chemistry, whereas low-viscosity **liquid-like** particles facilitate rapid gas–particle exchange and aging (Kuwata
60 and Martin, 2012; Berkemeier et al., 2014; Gou et al., 2025).

61 During the past decade, extensive laboratory studies have attempted to quantify aerosol viscosity using organic
62 or mixed organic–inorganic systems (Renbaum-Wolff et al., 2013a; Song et al., 2016b; Marshall et al., 2018;
63 Rovelli et al., 2019; Jeong et al., 2022; Tong et al., 2022; Mahant et al., 2023; Sheldon et al., 2023; Gou et al.,
64 2025). Measurements for SOA derived from anthropogenic (i.e., toluene, diesel fuel vapors, and phenolic
65 compounds) and biogenic (i.e., isoprene, α -pinene, β -ocimene, limonene, β -caryophyllene, **and** valencene)
66 precursors have revealed that viscosity can span more than ten orders of magnitude, from 10⁻³ Pa·s under humid
67 conditions to over $\sim 10^8$ Pa·s at low RH (Renbaum-Wolff et al., 2013b; Grayson et al., 2016; Hinks et al., 2016;
68 Song et al., 2016a; Song et al., 2019; Ullmann et al., 2019; Maclean et al., 2021; Smith et al., 2021; Baboomian
69 et al., 2022; Nikkho et al., 2024; Liu et al., 2026). These findings demonstrate that aerosol viscosity depends
70 strongly on chemical composition, including the organic-to-inorganic mass ratio (OIR), RH, and temperature.
71 However, almost all such data have been derived from laboratory-generated SOA or proxy mixtures, rather than
72 real ambient particles.

73 Seoul and Beijing represent typical urban environments influenced by mixed anthropogenic emissions, where
74 PM_{2.5} is frequently dominated by OA and secondary inorganic aerosols (SIA) (Son et al., 2012; Tao et al., 2017;

75 Kim et al., 2018; Zhou et al., 2020; Kim et al., 2022; Qiu et al., 2023; Cheng et al., 2024b; Daellenbach et al.,
76 2024). Field studies have shown that OA frequently contributes more than half of the PM_{2.5} mass in Seoul and
77 Beijing and comprises a mixture of POA (traffic, cooking, biomass, or coal/solid-fuel combustion) and SOA
78 formed from various precursors under a range of meteorological conditions (Sun et al., 2010; Hu et al., 2017; Kim
79 et al., 2017; Zhao et al., 2019; Qiu et al., 2023; Cheng et al., 2024b). In such OA-rich urban environments, the
80 phase state and viscosity of PM_{2.5} are expected to be strongly controlled by the properties of the organic fraction,
81 with direct implications for hygroscopic growth, gas–particle partitioning, and particle diffusion (Shiraiwa et al.,
82 2011; Davies and Wilson, 2015; Hosny et al., 2016; Tong et al., 2022). Consequently, direct constraints on the
83 phase state and viscosity of urban OA-containing PM_{2.5} remain essential for interpreting PM_{2.5} composition and
84 reducing uncertainties in process-level air-quality assessments.

85 In this study, PM_{2.5} samples were collected from the urban environments of Seoul and Beijing during autumn
86 2023. The samples were characterized by high organic mass fractions, representing organic-rich urban aerosols.
87 Using filter extracts, we investigated RH-dependent phase behavior and morphological evolution using optical
88 microscopy. We then quantitatively constrained aerosol viscosity at a temperature of ~290 K under experimentally
89 accessible RH conditions (< ~45 %) using the poke-and-flow technique. Finally, we compared the determined
90 viscosities with those reported in previous laboratory studies of organic–inorganic aerosol systems. By providing
91 direct quantitative viscosity data for field-collected PM_{2.5} in urban environments, this work advances our
92 understanding of the physicochemical behavior of ambient aerosols in the real atmosphere.

93 **2. Experimental and methods**

94 **2.1 Measurement sites and collection of PM_{2.5} samples**

95 PM_{2.5} sampling was conducted at an urban environmental monitoring station in Bulgwang-dong, Seoul (37.61°
96 N, 126.93° E) and the Changping campus of Peking University, Beijing (40.25°N, 116.19°E). Both sites are
97 located in densely populated metropolitan areas characterized by heavy traffic and significant industrial activity,

98 making them representative of typical urban environments. At each site, a total of three PM_{2.5} samples were
99 collected on quartz-fiber filters (8 × 10 in., Pall Corporation, 7204) over the sampling period between September
100 and October 2023. Each sample was collected from 10:00 to 09:00 local time using a high-volume sampler
101 operating at approximately 1000 L min⁻¹ (SIBATA HV-1000R, Japan). During the sampling period, the average
102 ambient RH and temperature were 69 ± 12 % and 290 ± 4 K at the Seoul site, and 54 ± 13 % and 293 ± 4 K at the
103 Beijing site, respectively.

104 After collection, filters were individually sealed in aluminum foil, placed in zip-lock bags, and stored at ~255
105 K to minimize evaporative loss of semi-volatile compounds and microbial degradation. All morphology and
106 viscosity experiments were conducted within ~1 month of collection to limit changes in PM_{2.5} chemical and
107 physical properties. For the morphology and viscosity experiments, PM_{2.5} material was recovered from each filter
108 using a 1:1 (v/v) methanol–water extraction procedure designed to capture both hydrophilic and hydrophobic
109 species, as detailed in Sect. S1. The resulting extract was then nebulized onto a hydrophobic glass substrate
110 (Hampton Research, Canada) using a nebulizer (MEINHARD, PerkinElmer, USA) to generate PM_{2.5} droplets.
111 Details of the methods used to determine chemical compositions are provided in Sect. S2 of the Supplementary
112 Information.

113 2.2 Observation of morphological change upon dehydration

114 To investigate morphological changes of the PM_{2.5} droplets on a hydrophobic substrate, optical microscopy was
115 employed following the approach used in previous studies (Ham et al., 2019; Jeong et al., 2022; Song et al., 2022;
116 Seong et al., 2024; Song et al., 2025). Briefly, PM_{2.5} droplets were first equilibrated at ~100 % RH and 290 ± 1 K
117 for ~20 min in an RH- and temperature-controlled flow-cell (TSA12Gi, Instec, USA), and then the RH was
118 reduced at a rate of 0.5 % RH min⁻¹ down to ~0 % RH. During typical experiments, the morphological evolution
119 of the droplets before, during, and post-poking was monitored via optical microscopy (Olympus BX43, 40×
120 objective, Japan) and recorded with a CCD camera (DigiRetina 16, Tucsen, China). The RH sensor (Sensirion,

121 **SHT C3, Switzerland**) within the flow-cell was calibrated at 290 K using deliquescence RH of K_2CO_3 (44 % RH),
122 NaCl (76 % RH), and $(\text{NH}_4)_2\text{SO}_4$ (80 % RH) (Winston and Bates, 1960), with an associated uncertainty of ± 1.5
123 %. RH control was achieved by adjusting the mixing ratio of dry N_2 and H_2O -saturated vapor at a constant total
124 flow rate of 500 sccm. The experimental temperature (290 K) was selected to closely reflect the average conditions
125 at the sampling sites, thereby ensuring environmental relevance.

126 **2.3 Poke-and-flow technique**

127 The poke-and-flow technique was employed to determine the viscosity of highly viscous $\text{PM}_{2.5}$ droplets on a
128 hydrophobic substrate within a temperature- and RH-controlled flow-cell (Renbaum-Wolff et al., 2013b; Grayson
129 et al., 2015; Song et al., 2015; Song et al., 2025). **In this technique, a micrometer-scale droplet (~ 20 – $40 \mu\text{m}$ in
130 diameter) is mechanically deformed by a fine needle; the subsequent relaxation of the deformed shape is governed
131 by the competition between surface tension (restoring force) and viscous resistance, allowing viscosity to be
132 quantified from the observed relaxation timescale in combination with fluid-dynamics simulations (Sect. 2.4). The
133 morphological evolution of the droplets before, during, and after poking was monitored using optical microscopy
134 (Olympus CKX53 with a $40\times$ objective, Japan) and recorded with a CCD camera (Hamamatsu, C11440-42U30,
135 Japan).**

136 For viscosity measurements, droplets were first equilibrated at ~ 100 % RH, then RH was decreased to ~ 40 %
137 at ~ 1 % RH min^{-1} . Poking was performed at RH levels of ~ 40 %, ~ 30 %, ~ 20 %, ~ 10 %, and ~ 0 % using a fine
138 needle (Jung Rim Medical Industrial, South Korea) mounted on a micromanipulator (Narishige, model MO-152,
139 Japan). Before each poking, droplets were conditioned for ~ 1 h at the target RH to ensure equilibration, consistent
140 with previous studies on semisolid particles (Kiland et al., 2023; Gerrebos et al., 2024; Ullah et al., 2026).
141 Following poking, a hole formed and gradually closed; the time required for the equivalent hole diameter to
142 decrease to 50 % of its initial value was defined as the experimental flow time, $\tau_{(exp, flow)}$ (see Sect. 2.4 for details).
143 **In typical experiments, one to five particles per sample were analyzed at each RH, and the total number of droplets**

144 analyzed per sample ranged from one to nine. At $RH > \sim 50\%$, the $PM_{2.5}$ droplets behaved as low-viscosity liquids
145 and hole closure occurred too quickly to be captured within the imaging frame rate; therefore, $\tau_{(exp, flow)}$ could not
146 be determined under these conditions.

147 **2.4 Fluid-dynamic simulations**

148 Viscosities were determined from $\tau_{(exp, flow)}$ using finite-element fluid flow simulations in COMSOL Multiphysics
149 (version 5.5), employing the Laminar Flow and Moving Mesh interfaces to model viscous droplet flow and droplet
150 geometry deformation during relaxation, respectively, following established methods (Renbaum-Wolff et al.,
151 2013b; Grayson et al., 2015; Song et al., 2015; Song et al., 2016a). In the simulations, the poked droplet was
152 represented as a half-torus geometry, with the inner and outer diameters determined from optical images acquired
153 after poking. Since the inner hole could exhibit irregular, non-axisymmetric shapes, its perimeter was traced from
154 the optical images, the enclosed area was calculated, and an equivalent circular diameter ($d = (4A/\pi)^{1/2}$) was used
155 to define the initial torus geometry and $\tau_{(exp, flow)}$. For each particle for which flow was observed, the dynamic
156 viscosity was iteratively adjusted until $\tau_{(model, flow)}$ agreed with $\tau_{(exp, flow)}$ to within $\sim 1\%$ (Song et al., 2016a).

157 The key physical parameters required for the simulations, along with their lower- and upper-bound values, are
158 summarized in Table 1. The slip length, which characterizes the degree of velocity slip at the fluid–solid interface,
159 was bounded between 5 nm and 10 μm based on literature values for fluid–solid interactions at hydrophobic
160 surfaces (Schnell, 1956; Churaev et al., 1984; Watanabe et al., 1999; Baudry et al., 2001; Cheng and Giordano,
161 2002; Tretheway and Meinhart, 2002; Jin et al., 2004; Joseph and Tabeling, 2005; Choi and Kim, 2006; Zhu et
162 al., 2012; Li et al., 2014). For density and surface tension, bounds were selected based on representative aerosol
163 types: the lower bound used properties of isoprene-derived SOA (density: 1.2 g cm^{-3} , Li et al., 2022; surface
164 tension: 17 mN m^{-1} , <https://www.chemspider.com/>), and the upper bound applied values for supersaturated
165 ammonium sulfate (AS) (density: 1.7 g cm^{-3} , <https://www.chemspider.com/>; surface tension: 95 mN m^{-1} ,
166 Mikhailov et al., 2024). Although the sampled $PM_{2.5}$ is predominantly organic, inorganic salts such as AS can

167 become highly concentrated under dry conditions and strongly inhibit particle flow, providing a conservative
 168 upper viscosity limit. Contact angles were determined from side-view optical images of representative droplets
 169 on the substrate using ImageJ (Grayson et al., 2015), with measured values ranging from 30° to 75°, reflecting
 170 variability in droplet size and composition across the analyzed samples. The overall uncertainty of approximately
 171 two orders of magnitude in the derived viscosity stems from the variability in these input parameters, with the slip
 172 length being the primary contributor (Grayson et al., 2015; Song et al., 2015).

173 At low RH, droplets exhibited brittle cracking without relaxation, indicating non-flowing behavior. If no
 174 recovery was observed over ~2 h, a lower-bound viscosity of $\sim 1 \times 10^8$ Pa·s was assigned, following established
 175 practice in poke-and-flow studies (Renbaum-Wolff et al., 2013b; Jeong et al., 2022; Gerrebos et al., 2024;
 176 Gerrebos et al., 2025).

177 Table 1. Summary of the key input physical parameters and boundary conditions used in the COMSOL
 178 Multiphysics simulations for the lower- and upper-bound viscosity calculations. R and r denote the geometric
 179 parameters of the torus: Here, R is the distance from the center of the hole to the midpoint of the torus ring, and r
 180 is the radius of the torus ring.

Parameter	Lower-bound calculation	Upper-bound calculation
Slip length*	5 nm	10 μ m
Density**	1.2 g cm ⁻³	1.7 g cm ⁻³
Surface tension***	17 mN m ⁻¹	95 mN m ⁻¹
Contact angle	30° ($r < 2R$), 75° ($r > 2R$)	75° ($r < 2R$), 30° ($r > 2R$)

* The slip length is based on values reported in the literature for fluid–solid interactions at hydrophobic surfaces (Schnell, 1956; Churaev et al., 1984; Watanabe et al., 1999; Baudry et al., 2001; Cheng and Giordano, 2002; Trethewey and Meinhart, 2002; Jin et al., 2004; Joseph and Tabeling, 2005; Choi and Kim, 2006; Zhu et al., 2012; Li et al., 2014). ** Values taken from Li et al. (2022) and <https://www.chemspider.com/>. *** Values taken from www.chemspider.com/ and Mikhailov et al. (2024).

181 3. Results and discussion

182 3.1 Chemical characteristics of PM_{2.5}

183 The chemical composition of PM_{2.5} collected from Seoul and Beijing during autumn 2023 is summarized in Fig.
184 S2 and Table 2. The major components include organic matter (OM), sulfate, nitrate, ammonium, and minor ions
185 (e.g., K⁺, Na⁺, Ca²⁺, Mg²⁺, and Cl⁻). OM consistently accounted for more than ~65 % of the total PM_{2.5} mass across
186 all the samples. Although this classification captures the dominant aerosol constituents, trace species such as
187 metals, black carbon, and crustal materials also contribute to PM_{2.5} but were not the focus of this study.

188 The daily PM_{2.5} concentrations at the Seoul and Beijing sites ranged from ~7.0 to 31.7 μg m⁻³, encompassing
189 both relatively clean and polluted conditions based on the World Health Organization (WHO) 24-hour air quality
190 guideline of 15 μg m⁻³ (World Health Organization, 2021). Over this range, the OIR varied from approximately
191 2:1 to 8:1, consistent with values commonly observed in various tropospheric environments (Hodzic et al., 2020;
192 Cheng et al., 2024a; Zhang et al., 2024).

193 The bulk O:C ratios (~0.4 – 0.5 at both sites, Table 2) fall within the typical range for urban OA (Aiken et al.,
194 2008; Chen et al., 2015; Zhou et al., 2020), indicating that the organics were overall moderately oxidized. This
195 composition suggests a substantial fraction of hydrophilic, highly oxygenated secondary organics alongside a non-
196 negligible pool of more hydrophobic, less oxidized material (Kim et al., 2025). Consequently, autumn-time OM
197 in Seoul and Beijing can be characterized as an amphiphilic mixture, consistent with previous mass spectrometric
198 observations of urban aerosols in the region (Kim et al., 2022). Although inorganic ions were not the dominant
199 mass component as reflected by the OIR ranges (Table 2), they still represented an important fraction of PM_{2.5},
200 primarily as SIA dominated by ammonium sulfate (AS) with additional contributions from ammonium nitrate
201 (AN) (Fig. S2; Sect. S2).

202 **Table 2.** Summary of mean ambient temperature, relative humidity (RH), mass concentration of PM_{2.5}, and
203 sulfate, nitrate, ammonium, and minor ions (K⁺, Na⁺, Ca²⁺, Mg²⁺, and Cl⁻) in Seoul and Beijing. The mean

204 concentration of organic material (OM) was determined by **subtracting** the inorganic salts, including sulfate,
 205 nitrate, ammonium, and minor ions, from the PM_{2.5} mass concentration. OIR refers to the organic-to-inorganic
 206 mass ratio, whereas O:C means oxygen-to-carbon elemental ratio. **The last column shows the total number of**
 207 **droplets analyzed by the poke-and-flow technique for each sample.**

Sampling date (mm/dd)	Ambient temperature (K)	Ambient RH (%)	PM _{2.5} μg m ⁻³	SO ₄ ²⁻ μg m ⁻³	NO ₃ ⁻ μg m ⁻³	NH ₄ ⁺ μg m ⁻³	Minor μg m ⁻³	OM μg m ⁻³	OIR	O:C	# of particles
Seoul											
09/30	291 ± 4	76 ± 6	16.8	2.2	0.6	0.9	0.3	12.9	3:1	0.45	7
10/12	290 ± 6	62 ± 13	16.4	1.9	1.0	1.0	0.2	12.3	3:1	0.47	9
10/15*	287 ± 4	84 ± 6	23.0	3.2	2.2	1.6	0.5	15.5	2:1	0.45	2
Beijing											
09/21	296 ± 2	61 ± 8	16.5	1.3	0.3	0.3	0.1	14.6	8:1	0.50	6
10/03	293 ± 3	61 ± 6	31.7	2.5	3.8	1.5	0.7	23.1	3:1	0.48	1
10/14*	289 ± 5	47 ± 15	7.0	1.0	0.7	0.3	0.3	4.7	3:1	0.52	3

208
 209 **3.2 Morphological characteristics and phase behavior of PM_{2.5} droplets**

210 To directly observe the phase behavior under progressively lowering RH, micrometer-sized droplets generated
 211 from PM_{2.5} extracts were monitored in a temperature- and RH-controlled flow-cell at 290 ± 1 K. This temperature
 212 closely matches the mean ambient conditions during the autumn sampling period at both sites (Seoul: 291 ± 6;
 213 Beijing: 291 ± 4 K; Table 2). Figure 1 shows optical images at four RH levels (~95 %, ~85 %, ~60 %, and ~20
 214 %), illustrating the evolution of particle morphology and phase state during dehydration. At high RH (~95 %),
 215 droplets behaved as a homogeneous **single-phase liquid**. They appeared smooth and rounded, with uniform optical
 216 contrast and no discernible internal structure, indicating that near water saturation, the urban PM_{2.5} extracts form
 217 well-mixed **single-phase liquid**.

218 As RH decreased to ~85 %, all droplets exhibited internal structuring consistent with liquid-liquid phase
 219 separation (LLPS) (Ciobanu et al., 2009; Song et al., 2012; Freedman, 2017; Ham et al., 2019; Lam et al., 2021;

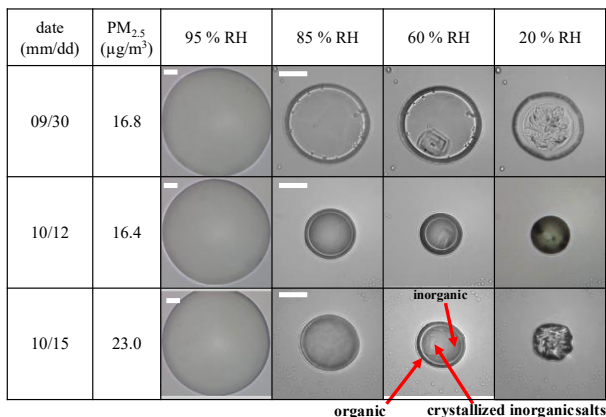
220 Freedman et al., 2024). Because the droplets remained single phase at ~95 % RH but had clearly phase-separated
221 by ~85 % RH, the observed separation RH in our experiments was constrained to between ~95 % and ~85 % RH.
222 Distinct inner and outer regions emerged, often accompanied by small inclusions or domains within the droplet.
223 These two-phase liquid morphologies adopted a core-shell geometry (Fig. 1), consistent with LLPS behavior
224 established in laboratory studies of mixed organic-inorganic particles and with the bulk composition of our
225 samples (O:C = 0.45–0.52; OIR = 2:1–8:1), for which LLPS is expected based on the O:C threshold of ~0.80
226 below which phase separation commonly occurs in organics mixed with inorganic salts such as AS or AN (Song
227 et al., 2013; You et al., 2013; Stewart et al., 2015; Kucinski et al., 2021; Huang et al., 2024).

228 Further dehydration to ~60 % RH frequently produced three-phase morphologies. In this regime, solid-like
229 domains coexisted with two liquid regions. Some solid-like inclusions appeared angular (Fig. 1), suggesting
230 crystallized inorganic salts or less soluble organic materials. Based on the bulk ionic composition (dominated by
231 secondary inorganic salts such as AS; Fig. S2 and Sect. S2), it is plausible that at least part of the solid-like material
232 was inorganic-rich, though contributions from non-crystalline organic-rich phases cannot be excluded. The
233 relative volumes of the inner liquid, outer shell, and crystalline regions varied among droplets, reflecting
234 compositional differences and drying history. These observations align with recent laboratory studies showing
235 that mixed organic-inorganic aerosols can exhibit complex multiphase behavior across wide RH ranges (Huang
236 et al., 2021).

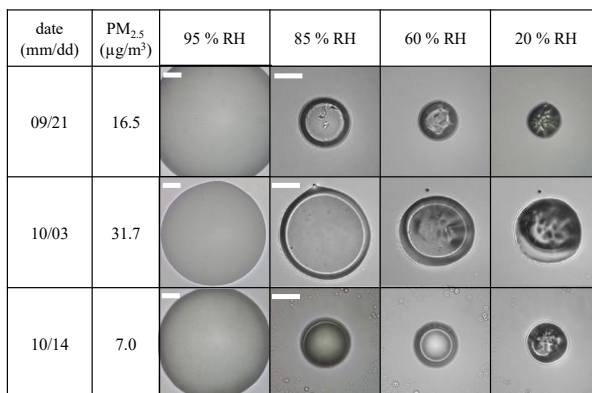
237 At low RH (~20 %), most droplets transitioned to non-liquid morphologies. Droplets displayed features
238 characteristic of efflorescence, either confined to the inner region or involving nearly the entire particle (Fig. 1).
239 These observations indicate that at least one phase had effectively lost its ability to flow, consistent with the
240 presence of effloresced inorganic solids and/or highly viscous organic material. Because morphology alone cannot
241 unambiguously distinguish crystalline solids from extremely viscous amorphous semisolids, these low-RH states

242 are treated as non-liquid. Quantitative viscosity of the bulk of PM_{2.5} is provided by poke-and-flow measurements
 243 in Sect. 3.3.

(a) Seoul



(b) Beijing



244

245

246 **Figure 1. Optical images obtained during RH decrease for (a) Seoul and (b) Beijing PM_{2.5} droplets showing phase**
 247 **separation, as RH decreases from ~95 % to 85, 60, and 20 %. Upon dehydration, particles transition from a**
 248 **homogeneous single-phase liquid to a core-shell morphology, illustrating separation of organic and inorganic**
 249 **components driven by water loss. The images at ~95 % RH are shown at the same scale, while those at ~85 % RH and**
 250 **lower are presented at a consistent scale to facilitate comparison between samples. The scale bar represents 20 μm.**

251 **Seoul (10/15) and Beijing (10/14) samples have already been reported by Song et al. (2025) and are included here for**
252 **completeness of discussion.**

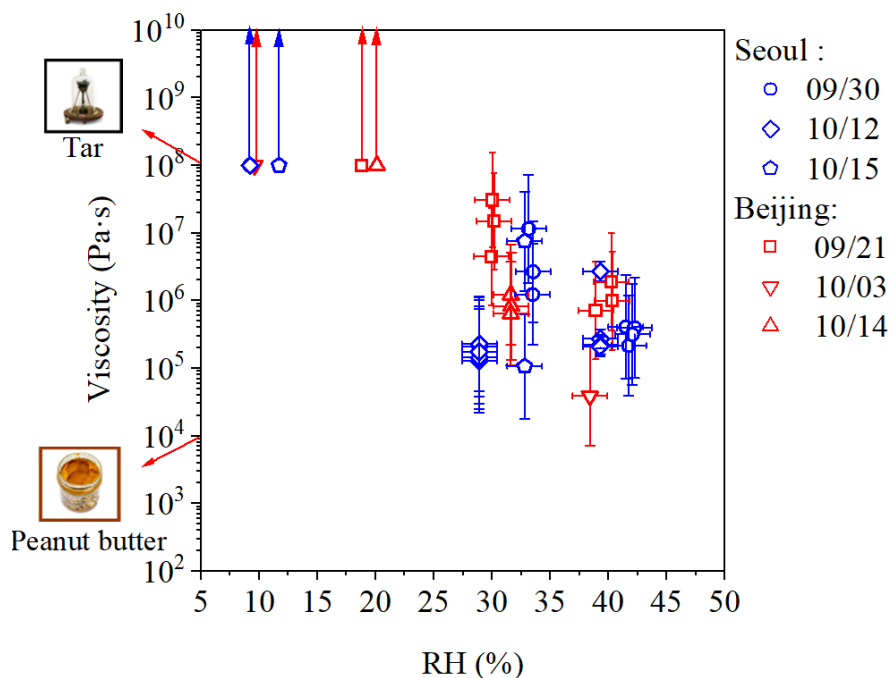
253 **3.3 Viscosity and phase state of organic-rich PM_{2.5}**

254 To quantitatively constrain the RH-dependent viscosity of the bulk of the organic-rich urban PM_{2.5}, poke-and-
255 flow experiments were conducted, and the resulting $\tau_{(exp, flow)}$ values (Fig. S1) were analyzed in combination with
256 fluid-dynamic simulations using COMSOL Multiphysics. Representative optical images of the poke-and-flow
257 experiments are shown in Fig. 3. Previous field studies of ambient PM_{2.5} have primarily focused on qualitative
258 phase-state classifications (e.g., liquid, semisolid, or solid) (Bateman et al., 2016; Song et al., 2022; Meng et al.,
259 2024; Seong et al., 2024). However, quantitative, RH-resolved viscosity measurements for urban aerosols remain
260 scarce.

261 Figure 2 shows the viscosities of PM_{2.5} droplets collected from Seoul and Beijing within the experimentally
262 accessible RH range (RH < ~45 %). Across the RH range between ~45 and 25 %, the mean viscosities were
263 approximately 10⁴–10⁸ Pa·s, corresponding to consistencies ranging from peanut butter to tar pitch (Koop et al.,
264 2011; Reid et al., 2018). The autumn-time urban PM_{2.5} droplets examined here predominantly fell within the
265 semisolid regime in the studied RH ranges.

266 Although the poke-and-flow experiments were conducted under drier conditions (RH < ~45 %) than the mean
267 ambient RH during sampling (Seoul: 69 ± 12 %; Beijing: 54 ± 13 %), direct quantitative viscosity constraints at
268 higher RH could not be obtained, as the droplets behaved as low-viscosity liquids above ~45 % RH and relaxed
269 too rapidly to yield a resolvable $\tau_{(exp, flow)}$. Nevertheless, comparison with sucrose–AS–H₂O systems (Fig. 5)
270 indicates that semisolid behavior may still occur during at least episodic portions of the sampling period,
271 particularly for the Beijing samples and during lower-RH periods in Seoul. These results, therefore, suggest that
272 semisolid behavior of urban PM_{2.5} cannot be ruled out even under the RH ambient conditions observed during the
273 sampling period, particularly during drier episodes, and that viscosity measurements at higher RH remain an
274 important target for future work. The derived viscosities exhibited substantial sample-to-sample and droplet-to-

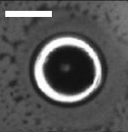
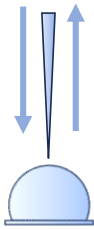
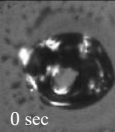
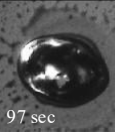
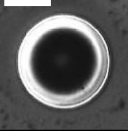
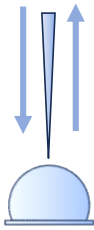
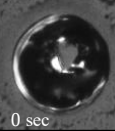
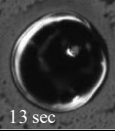
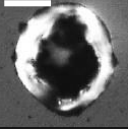
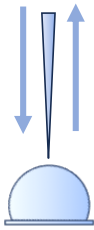
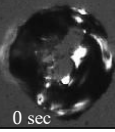
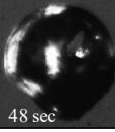
275 droplet variability. The observed spread in $\tau_{(exp,flow)}$ between individual droplets from the same filter extract likely
 276 reflects variability in local phase state upon dehydration, differences in the RH threshold at which inorganic salts
 277 become supersaturated, and minor experimental factors such as droplet size heterogeneity, all of which can
 278 strongly influence viscosity at intermediate RH.



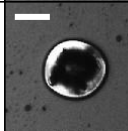
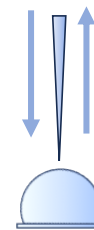
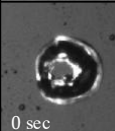
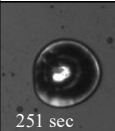
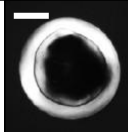
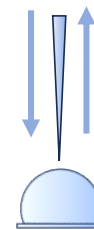
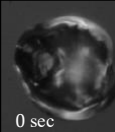
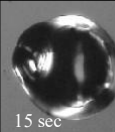
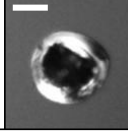
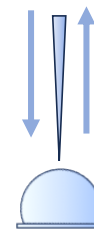
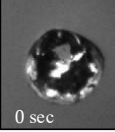
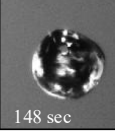
279
 280 **Figure 2.** Mean viscosities of PM_{2.5} droplets derived from experimentally measured flow times (Fig. S1) using poke-
 281 and-flow measurements and COMSOL simulations. Markers denote mean values, with y-error bars indicating upper
 282 and lower bound deviations from the mean, calculated as the difference between the mean and the corresponding upper
 283 and lower bounds derived from simulations using minimum and maximum input parameters (Table 1). Upward arrows
 284 indicate lower-limit viscosities where no restorative flow was observed within the experimental timescale. The x-axis
 285 error bars represent the RH sensor uncertainty (±1.5 %), as determined from our RH sensor calibration in the flow-
 286 cell (Sect. 2.2). Reference viscosities for peanut butter (~10⁴ Pa·s) and tar (~10⁸ Pa·s) are shown for comparison (Koop
 287 et al., 2011; Reid et al., 2018).

288 At lower RH, PM_{2.5} droplets frequently exhibited brittle cracking without observable relaxation (Fig. 4).
289 When no restorative flow was detected over observation periods exceeding two hours, a conservative lower-limit
290 viscosity of $\sim 10^8$ Pa·s was assigned, consistent with the practical lower limit that can be constrained using the
291 poke-and-flow technique (Renbaum-Wolff et al., 2013b; Grayson et al., 2015; Jeong et al., 2022; Gerrebos et al.,
292 2024). Under these conditions, PM_{2.5} droplets from Seoul exhibited cracking at RH values of ~ 9.2 %, ~ 9.2 %, and
293 ~ 11.7 %, whereas droplets from Beijing cracked at comparatively higher and more variable RH values of ~ 18.8
294 %, ~ 9.6 %, and ~ 20.1 % on each date. These observations indicate that the RH threshold for the transition to non-
295 flowing behavior is dependent on PM_{2.5} composition. The inferred lower-bound viscosities correspond to
296 consistencies comparable to, or exceeding, those of tar-like materials, suggesting extremely viscous properties or
297 arrested internal flow under dry conditions.

(a) Seoul

Date (mm/dd)	PM _{2.5} (μg/m ³)	RH	Pre-poking	Poking	First frame post-poking	$\tau_{(exp, flow)}$ frame post-poking
09/30	16.8	~ 40 %			 0 sec	 97 sec
10/12	16.4	~ 40 %			 0 sec	 13 sec
10/15	23.0	~ 33 %			 0 sec	 48 sec

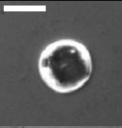
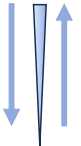
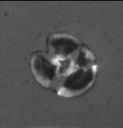
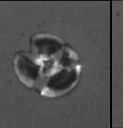
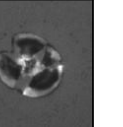
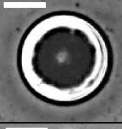


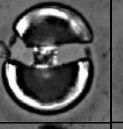

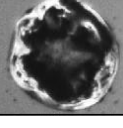
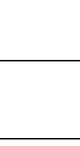
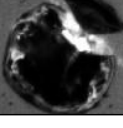
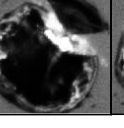
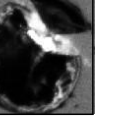
(b) Beijing

Date (mm/dd)	PM _{2.5} (μg/m ³)	RH	Pre-poking	Poking	First frame post-poking	$\tau_{(exp, flow)}$ frame post-poking
09/21	16.5	~ 40 %			 0 sec	 251 sec
10/03	31.7	~ 40 %			 0 sec	 15 sec
10/14	7.0	~ 31 %			 0 sec	 148 sec

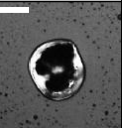

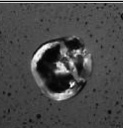
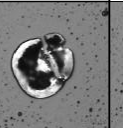
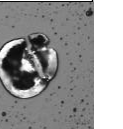
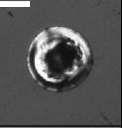

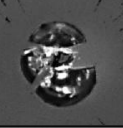
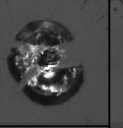
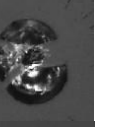
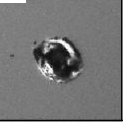
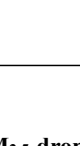
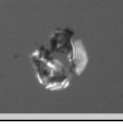
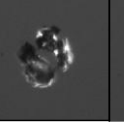

298

299 **Figure 3. Poke-and-flow results for (a) Seoul and (b) Beijing PM_{2.5} droplets at ~40 – ~30 % RH conditions under which**
 300 **particle flow was observed. The first post-poking frame corresponds to the image taken immediately after needle**
 301 **retraction (t = 0 sec), and the later frame corresponds to the experimental flow time, $\tau_{(exp, flow)}$, when the inner hole**
 302 **diameter has decreased to 50 % of its initial size. The scale bar represents 20 μm.**

(a) Seoul

date (mm/dd)	PM _{2.5} (μg/m ³)	RH	pre-poking	poking	after 0 sec	after 1 hr	after 2 hrs
09/30	16.8	~9.2 %					
10/12	16.4	~9.2 %					
10/15	23.0	~11.7 %					

(b) Beijing

date (mm/dd)	PM _{2.5} (μg/m ³)	RH	pre-poking	poking	after 0 sec	after 1 hr	after 2 hrs
09/21	16.5	~18.8 %					
10/03	31.7	~9.6 %					
10/14	7.0	~20.1 %					

303

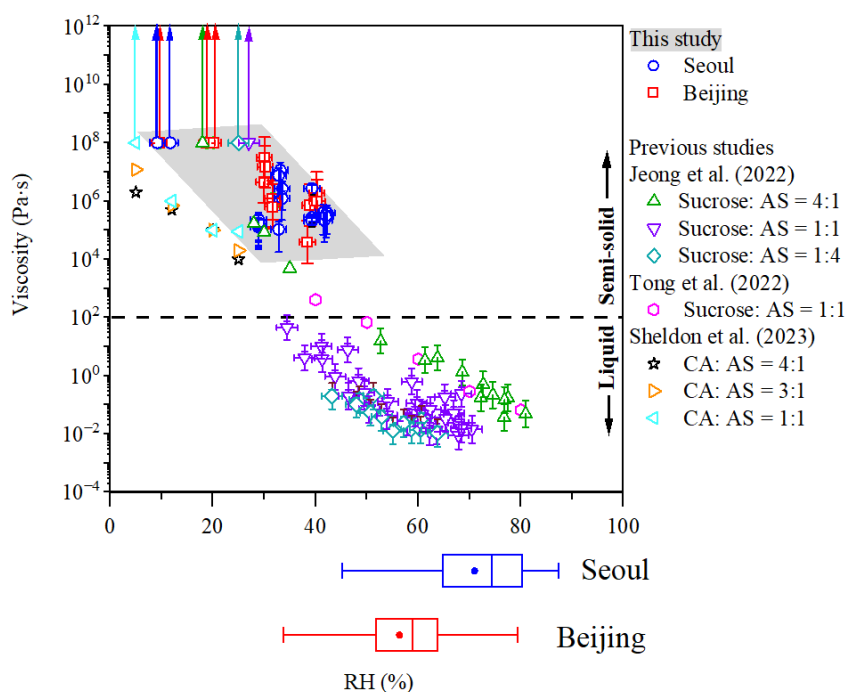
304 **Figure 4. Optical images of (a) Seoul and (b) Beijing PM_{2.5} droplets exhibiting cracking at their respective RH after**
305 **poking. The scale bar represents 20 μm.**

306 The autumn PM_{2.5} samples examined here were overall organic-rich, with sulfate as the dominant inorganic
307 ion, primarily present as AS (Sect. 3.1). The RH-resolved viscosities measured for PM_{2.5} were compared with
308 laboratory measurements for internally mixed organic–AS systems, commonly used as surrogates for organic-

309 rich, sulfate-containing aerosols (Fig. 5). Previous laboratory studies indicated that citric acid (CA)–AS–H₂O
310 systems exhibited relatively low viscosities and weak RH dependence at comparable RH, whereas sucrose–AS–
311 H₂O systems showed a pronounced increase in viscosity upon dehydration, reaching $\sim 10^3$ – 10^5 Pa·s at RH of ~ 20 –
312 30 % and approaching $\sim 10^8$ Pa·s upon cracking at lower RH (e.g., sucrose–AS–H₂O for 4:1 and 1:1, Fig. 5) (Jeong
313 et al., 2022; Tong et al., 2022; Sheldon et al., 2023). Accordingly, sucrose–AS systems represent the highest RH-
314 dependent viscosities reported among commonly used laboratory surrogates, providing an appropriate upper
315 reference for comparison with organic-rich urban PM_{2.5}.

316 Within the experimentally accessible RH range ($< \sim 45$ %) in this study using the poke-and-flow technique,
317 the viscosities determined for organic-rich urban PM_{2.5} droplets were comparable to the highest values reported
318 for sucrose–AS laboratory systems and, in several cases, exceeded this upper range, while remaining consistently
319 higher than those reported for CA–AS systems at comparable RH. These results suggest that organic-rich urban
320 PM_{2.5} can attain viscosities at least as high as those of the most viscous laboratory surrogate systems commonly
321 used in aerosol research.

322 Our conclusions are based on a limited number of filters and droplets, and the experiments were conducted on
323 micrometer-sized extracted droplets on a substrate, which may not fully represent submicron ambient particles.
324 Future studies extending viscosity measurements to a larger number of samples across different seasons and to
325 smaller, atmospherically relevant particle sizes would further constrain the phase behavior of urban PM_{2.5} under
326 real atmospheric conditions.



327

328 **Figure 5.** RH-dependent viscosities of Beijing and Seoul PM_{2.5} droplets compared with sucrose–AS–H₂O and citric acid
 329 (CA)–AS–H₂O systems from previous studies (Jeong et al., 2022; Tong et al., 2022; Sheldon et al., 2023). **Viscosities in**
 330 **the previous studies were determined using the poke-and-flow technique (Jeong et al., 2022), a dual optical tweezer**
 331 **system (Tong et al., 2022), and the droplet coalescence method (Sheldon et al., 2023).** y-error bars in this study indicate
 332 uncertainty ranges of the modeled viscosities for data corresponding to Fig. 2. Box plots in the lower panel show hourly
 333 RH distributions in Seoul and Beijing derived from the days on which PM_{2.5} samples were collected, with markers
 334 indicating mean values, boxes representing the 25th–75th percentiles, and whiskers showing minimum and maximum
 335 values.

336 4. Conclusion

337 In this study, we investigated the phase behavior and viscosity of organic-rich urban PM_{2.5} collected during
 338 autumn from Seoul and Beijing, using filter extracts. Optical microscopy observations qualitatively revealed RH-
 339 driven phase transitions in all analyzed samples during dehydration, including well-mixed **single-phase liquid**,

340 two-phase liquid and three-phases, followed by the development of non-flowing morphologies at lower RH. Using
341 the poke-and-flow technique coupled with fluid-dynamic simulations, we quantitatively constrained aerosol
342 viscosity at ~290 K within the experimentally accessible RH range ($RH < \sim 45\%$). For ~45–25 % RH, the inferred
343 viscosities of organic-rich $PM_{2.5}$ spanned approximately 10^4 – 10^8 Pa·s, corresponding to semisolid to non-flowing
344 behavior on the experimental timescale. At lower RH, brittle cracking without observable relaxation was
345 observed, and conservative lower-limit viscosities of $\sim 10^8$ Pa·s were assigned.

346 When placed in the context of existing laboratory studies, the viscosities inferred for organic-rich urban $PM_{2.5}$
347 were comparable to the highest values reported for sucrose–AS–H₂O laboratory systems and, in several cases,
348 exceeded this upper range within the RH interval of 20–45 %, while remaining consistently higher than those
349 reported for CA–AS systems at similar RH. These findings demonstrated that organic-rich urban $PM_{2.5}$ can attain
350 viscosities at the upper end of RH-dependent viscosity values previously reported for laboratory-generated
351 organic–inorganic aerosol surrogates. Although these constraints were derived from micrometer-sized filter
352 extract droplets and therefore do not fully preserve the native morphology or size of ambient submicron particles,
353 they provide direct, field-based quantitative benchmarks under atmospherically relevant low-RH conditions. Such
354 highly viscous states are expected to slow intra-particle diffusion and inhibit internal mixing, with implications
355 for gas–particle partitioning and multiphase chemical processing in urban aerosols. However, these conclusions
356 are based on a limited number of filter samples and droplets, and future studies extending measurements across
357 additional seasons, compositions, and particle sizes will be essential for further constraining the role of aerosol
358 viscosity in urban atmospheric processes.

359 **Author contributions**

360 Mijung Song: Conceptualization, Data curation, Funding acquisition, Software, Supervision, Writing–review &
361 editing. Atta Ullah: Data curation, Software, Writing–original draft, Writing–review & editing. Ji Yi Lee: Data
362 curation, Review & editing. Kyoung-Soon Jang: Data curation, Review & editing. Zhijun Wu: Data curation,
363 Review & editing.

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369 **Notes**

370 The authors declare no competing financial interest.

371 **Acknowledgment**

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373

374

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