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Highly Viscous Phase Behavior of Organic-Rich Urban $PM_{2.5}$

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Abstract

Atmospheric aerosol viscosity strongly influences particle phase state, internal mixing, and multiphase chemical processes, yet direct quantitative constraints for ambient urban $PM_{2.5}$ remain limited. Here, we investigated the phase behavior and viscosity of organic-rich $PM_{2.5}$ samples collected during autumn 2023 from the urban environments of Seoul and Beijing. Using filter extracts, relative humidity (RH)-dependent phase transitions and morphological evolution of the droplets were examined by optical microscopy, revealing frequent two-phase and three-phase morphologies during dehydration. Aerosol viscosity was quantitatively constrained at ~290 K under experimentally accessible RH condition (≤ ~40%) using the poke-and-flow technique coupled with fluid-dynamic simulations, yielding viscosities spanning from ~10^4 to > ~10^8 Pa·s. Compared with previously reported laboratory-based viscosity measurements, the inferred viscosities of ambient $PM_{2.5}$ were comparable to or exceeded those reported for organic-rich ternary systems (i.e., sucrose–AS–H_2O), that are commonly used as laboratory proxy systems in aerosol viscosity studies. These results indicate that organic-rich urban $PM_{2.5}$ can exhibit highly viscous, semisolid to non-flowing behavior and provide quantitative, field-based, compositionally constrained viscosity data for urban aerosols.

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

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1. Introduction

Atmospheric fine particulate matter ($PM_{2.5}$) in urban megacities undergoes a range of general physicochemical processes, including hygroscopic growth, phase transitions, viscosity changes, and diffusion-limited internal mixing, which are strongly modulated by relative humidity (RH) (Swietlicki et al., 2008; Guo et al., 2014; Reid et al., 2018; Song et al., 2022; Freedman et al., 2024; Tan et al., 2024). These processes play a critical role in gas–particle partitioning (Zuend and Seinfeld, 2012; Zhou et al., 2013; Gkatzelis et al., 2018; Zaveri et al., 2020), heterogeneous chemical reactivity (Li et al., 2020; Rasool et al., 2023; Song et al., 2025), aging dynamics (Shiraiwa et al., 2011; Liu et al., 2025), and cloud nucleation potential (Suda et al., 2014; Cheung et al., 2020; Pöhlker et al., 2023), yet their underlying mechanisms remain poorly constrained by empirical observations. This limitation fundamentally undermines the accurate parametrization of urban aerosol impacts within the regional and global atmospheric models.

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

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Among the physical properties affected by the chemical complexity and atmospheric evolution of $PM_{2.5}$, viscosity is particularly important for controlling particle phase state and reactivity. Viscosity is the internal resistance of a material to flow or deformation, quantifying the molecular mobility within a condensed phase. In the atmospheric context, viscosity serves as a critical indicator of particle phase state: aerosols with viscosities below 10^2 Pa·s behave as liquids, those between 10^2 and 10^{12} Pa·s are semisolids, and particles exceeding 10^{12} Pa·s are solids (Koop et al., 2011; Zobrist et al., 2011). Variations in viscosity can dictate how rapidly molecules diffuse, mix, and react within aerosol particles. A highly viscous particle restricts internal diffusion and slows multiphase chemistry, whereas low-viscosity liquid particles facilitate rapid gas–particle exchange and aging (Kuwata and Martin, 2012; Berkemeier et al., 2014; Gou et al., 2025).

During the past decade, extensive laboratory studies have attempted to quantify aerosol viscosity using organic or mixed organic–inorganic systems (Renbaum-Wolff et al., 2013a; Song et al., 2016b; Freedman, 2017; Marshall et al., 2018; Rovelli et al., 2019; Jeong et al., 2022; Tong et al., 2022; Mahant et al., 2023; Sheldon et al., 2023; Gou et al., 2025). Measurements for SOA derived from anthropogenic (i.e., toluene, diesel fuel vapors, and phenolic compounds) and biogenic (i.e., isoprene, α-pinene, β-ocimene, limonene, β-caryophyllene, valencene, and farnesene isomer mixture) precursors have revealed that viscosity can span more than ten orders of magnitude, from 10^{-3} Pa·s under humid conditions to over ~10^8 Pa·s at low RH (Renbaum-Wolff et al., 2013b; Grayson et al., 2016; Hinks et al., 2016; Song et al., 2016a; Song et al., 2019; Ullmann et al., 2019; Maclean et al., 2021; Smith et al., 2021; Nikkho et al., 2024; Liu et al., 2026). These findings demonstrate that aerosol viscosity depends strongly on chemical composition, including the organic-to-inorganic mass ratio (OIR), RH, and temperature. However, almost all such data have been derived from laboratory-generated SOA or proxy mixtures, rather than real ambient particles.

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

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

In this study, $PM_{2.5}$ samples were collected from the urban environments of Seoul and Beijing during autumn 2023. The samples were characterized by high organic mass fractions, representing organic-rich urban aerosols. Using filter extracts, we investigated RH-dependent phase behavior and morphological evolution using optical microscopy. We then quantitatively constrained aerosol viscosity at a temperature of ~290 K under experimentally accessible RH conditions (≤ ~40%) using the poke-and-flow technique. Finally, we compared the determined viscosities with those reported in previous laboratory studies of organic–inorganic aerosol systems. By providing direct quantitative viscosity data for field-collected $PM_{2.5}$ in urban environments, this work advances our understanding of the physicochemical behavior of ambient aerosols in the real atmosphere.

2. Experimental and methods

2.1 Measurement sites and collection of $PM_{2.5}$ samples

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

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making them representative of typical urban environments. At each site, a total of three $PM_{2.5}$ samples were collected on quartz-fiber filters (8 × 10 in., Pall Corporation, 7204) over the sampling period between September and October 2023. Each sample was collected from 10:00 to 09:00 local time using a high-volume sampler operating at approximately 1000 L min^{-1} (SIBATA HV-1000R, Japan). During the sampling period, the average ambient RH and temperature were 69 ± 12 % and 290 ± 4 K at the Seoul site, and 54 ± 13 % and 293 ± 4 K at the Beijing site, respectively. After collection, filters were stored at a temperature of ~255 K for subsequent analysis within ~1 month. For the morphology (Sect. 2.2) and viscosity (Sect. 2.3) experiments, $PM_{2.5}$ droplets prepared from each $PM_{2.5}$ filter extract, as detailed in Sect. S2, were deposited onto a hydrophobic glass substrate (Hampton Research, Canada) using a nebulizer (MEINHARD, PerkinElmer, USA). Details of the methods used to determine chemical compositions are provided in Sect. S1 of the Supplementary Information. Table 1 also summarizes the sampling information and the chemical composition of the six $PM_{2.5}$ samples.

2.2 Observation of morphological change upon dehydration

To investigate morphological changes of the $PM_{2.5}$ droplets on a hydrophobic substrate, optical microscopy was employed following the approach used in previous studies (Ham et al., 2019; Jeong et al., 2022; Song et al., 2022; Seong et al., 2024; Song et al., 2025). In brief, before the experiment, the $PM_{2.5}$ droplets were equilibrated at ~100% RH and 290 ± 1 K for ~20 min, and then the RH was reduced at a rate of 0.5 % RH min^{-1} in an RH and temperature-controlled flow cell (TSA12Gi, Instec, USA) down to ~0% RH. During typical experiments, the morphological evolution of the droplets before, during, and after poking was monitored via optical microscopy (Olympus BX43, 40× objective, Japan) and recorded with a CCD camera (DigiRetina 16, Tucsen, China). The RH sensor within the flow cell was calibrated at 290 ± 1 K using deliquescence relative humidity of K_2CO_3 (44% RH), NaCl (76% RH), and $(NH_4)_2SO_4$ (80% RH), with an associated uncertainty of ±1.5%. RH control was achieved by adjusting the mixing ratio of dry N_2 and saturated H_2O vapor, while maintaining a constant total flow

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rate of 500 sccm. The experimental temperature (290 ± 1 K) was selected to closely reflect the average conditions at the sampling sites, thereby ensuring environmental relevance.

2.3 Poke-and-Flow technique coupled with fluid simulations

The poke-and-flow technique was employed to determine the viscosity of highly viscous $PM_{2.5}$ droplets on a hydrophobic substrate within a temperature- and RH-controlled flow cell (Grayson et al., 2015; Song et al., 2015; Song et al., 2025). Droplets (~20–40 μm in diameter) prepared from $PM_{2.5}$ filter extract were deposited onto a hydrophobic glass substrate and conditioned in the flow-cell. Their morphological evolution before, during, and after poking was monitored using optical microscopy (Olympus CKX53 with a 40× objective, Japan) and recorded with a CCD camera (Hamamatsu, C11440-42U30, Japan). For viscosity measurements, droplets were first equilibrated at ~100% RH, then RH was decreased to ~40% at ~1% RH min^{-1}. Poking was performed at RH levels of ~40%, ~30%, ~20%, ~10%, and ~0% using a fine needle (Jung Rim Medical Industrial, South Korea) mounted on a micromanipulator (Narishige, model MO-152, Japan) to identify the critical RH for particle cracking. Before each poking, droplets were conditioned for ~1 h at the target RH to ensure equilibration (Sect. S4), consistent with previous studies on semisolid particles (Kiland et al., 2023; Gerrebos et al., 2024). Following poking, a hole formed and gradually closed; the time required for the hole to recover 50% of its initial diameter was defined as the experimental flow time, $\tau_{(exp,\ flow)}$. In typical experiments, one to five particles per sample were analyzed at each RH. At RH > ~40%, the $PM_{2.5}$ droplets behaved as low-viscosity liquids and rapidly relaxed upon poking, resulting in immediate hole closure and the absence of a well-defined $\tau_{(exp,\ flow)}$. As a result, the poke-and-flow method was not suitable for quantitatively constraining viscosity under these high-RH conditions.

Viscosities were then determined from $\tau_{(exp,\ flow)}$ using finite-element fluid flow simulations in COMSOL Multiphysics, following established methods (Renbaum-Wolff et al., 2013b; Grayson et al., 2015; Song et al., 2015; Song et al., 2016a) detailed in Sect. S3. Model parameters, including slip length, surface tension, contact angle, and density, are summarized in Table S2. Viscosity was iteratively adjusted until the simulated flow time

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$\tau_{(model,\ flow)}$ matched $\tau_{(exp,\ flow)}$. At low RH, droplets exhibited brittle cracking without relaxation, indicating non-flowing behavior. If no recovery occurred over 2 h, a lower-bound viscosity of $\sim 1 \times 10^8$ Pa·s was assigned (Renbaum-Wolff et al., 2013b; Jeong et al., 2022; Gerrebos et al., 2024; Gerrebos et al., 2025).

3. Results and discussion

3.1 Chemical characteristics of $PM_{2.5}$

The chemical composition of $PM_{2.5}$ collected from Seoul and Beijing during autumn 2023 is summarized in Fig. S2 and Table 1. The major components include organic matter (OM), sulfate, nitrate, ammonium, and minor ions (e.g., K^+, Na^+, Ca^{2+}, Mg^{2+}, and Cl^-). OM consistently accounted for more than ~65% of the total $PM_{2.5}$ mass across all the samples. Although this classification captures the dominant aerosol constituents, trace species such as metals, black carbon, and crustal materials also contribute to $PM_{2.5}$ but were not the focus of this study.

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

The bulk O:C ratios (~0.4 – 0.5 at both sites, Table 1) fall within the typical range for urban OA (Aiken et al., 2008; Chen et al., 2015; Zhou et al., 2020), indicating that the organics were overall moderately oxidized. This composition suggests a substantial fraction of hydrophilic, highly oxygenated secondary organics alongside a non-negligible pool of more hydrophobic, less oxidized material (Kim et al., 2025). Consequently, autumn-time OM in Seoul and Beijing can be characterized as an amphiphilic mixture, consistent with previous mass spectrometric observations of urban aerosols in the region (Kim et al., 2022). Although inorganic ions were not the dominant mass component as reflected by the OIR ranges (Table 1), they still represented an important fraction of $PM_{2.5}$,

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primarily as SIA dominated by ammonium sulfate (AS) with additional contributions from ammonium nitrate (AN) (Fig. S2; Sect. S1).

Table 1. Summary of mean ambient temperature, relative humidity (RH), mass concentration of $PM_{2.5}$, and sulfate, nitrate, ammonium, and minor ions (K^+, Na^+, Ca^{2+}, Mg^{2+}, and Cl^-) in Seoul and Beijing. The mean concentration of organic material (OM) was determined by deducing the inorganic components, including sulfate, nitrate, ammonium, and minor ions, from the $PM_{2.5}$ mass concentration. OIR refers to the organic-to-inorganic mass ratio, whereas O:C means oxygen-to-carbon elemental ratio.

Sampling date	Ambient temperature	Ambient RH	$PM_{2.5}$	SO_4^{2-}	NO_3^-	NH_4^+	Minor	OM	OIR	O:C
(mm/dd)	(K)	(%)	$\mu g/m^{-3}$	$\mu g/m^{-3}$	$\mu g/m^{-3}$	$\mu g/m^{-3}$	$\mu g/m^{-3}$	$\mu g/m^{-3}$		
Seoul										
09/30	291 ± 4	76 ± 6	16.8	2.2	0.6	0.9	0.3	12.9	3:1	0.45
10/12	290 ± 6	62 ± 13	16.4	1.9	1.0	1.0	0.2	12.3	3:1	0.47
10/15*	287 ± 4	84 ± 6	23.0	3.2	2.2	1.6	0.5	15.5	2:1	0.45
Beijing										
09/21	296 ± 2	61 ± 8	16.5	1.3	0.3	0.3	0.1	14.6	8:1	0.50
10/03	293 ± 3	61 ± 6	31.7	2.5	3.7	1.5	0.7	23.1	3:1	0.48
10/14*	289 ± 5	47 ± 15	7.0	1.0	0.7	0.3	0.3	4.7	3:1	0.52

*Data from Song et al. (2025).

3.2 Morphological characteristics and phase behavior of $PM_{2.5}$ droplets

To directly observe the phase behavior under progressively lowering RH, micrometer-sized droplets generated from $PM_{2.5}$ extracts were monitored in a temperature- and RH-controlled flow-cell at 290 ± 1 K. This temperature closely matches the mean ambient conditions during the autumn sampling period at both sites (Seoul: 291 ± 6; Beijing: 291 ± 4 K; Table 1). Figure 1 shows optical images at four RH levels (~95 %, ~85 %, ~60 %, and ~20 %), illustrating the evolution of particle morphology and phase state during dehydration. At high RH (~95 %), droplets behaved as homogeneous single-liquid particles. They appeared smooth and rounded, with uniform

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optical contrast and no discernible internal structure, indicating that near water saturation, the urban $PM_{2.5}$ extracts form well-mixed single liquid droplets.

As RH decreased to ~85 %, all droplets exhibited internal structuring consistent with liquid-liquid phase separation (LLPS) (Ciobanu et al., 2009; Song et al., 2012; Freedman, 2017; Ham et al., 2019; Lam et al., 2021; Freedman et al., 2024). Distinct inner and outer regions emerged, often accompanied by small inclusions or domains within the droplet. These two-liquid morphologies typically adopted a core–shell geometry (Fig. 1), consistent with LLPS criteria established in laboratory studies of mixed organic–inorganic particles, where phase separation commonly occurs when moderately oxidized organics are mixed with inorganic salts such as AS or AN (Song et al., 2013; You et al., 2013; Stewart et al., 2015; Kucinski et al., 2021; Huang et al., 2024).

Further dehydration to ~60% RH frequently produced three-phase morphologies. In this regime, solid-like domains coexisted with two liquid regions. Some solid-like inclusions appeared angular (Fig. 1), suggesting crystallized inorganic salts or less soluble organic materials. Based on the bulk ionic composition (dominated by secondary inorganic salts such as AS; Fig. S2 and Sect. S1), it is plausible that at least part of the solid-like material was inorganic-rich, though contributions from non-crystalline organic-rich phases cannot be excluded. The relative volumes of the inner liquid, outer shell, and crystalline regions varied among droplets, reflecting compositional differences and drying history. These observations align with recent laboratory studies showing that mixed organic–inorganic aerosols can exhibit complex multiphase behavior across wide RH ranges (Huang et al., 2021).

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

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qualitatively as non-liquid. Quantitative viscosity constraints are provided by poke-and-flow measurements in Sect. 3.3.

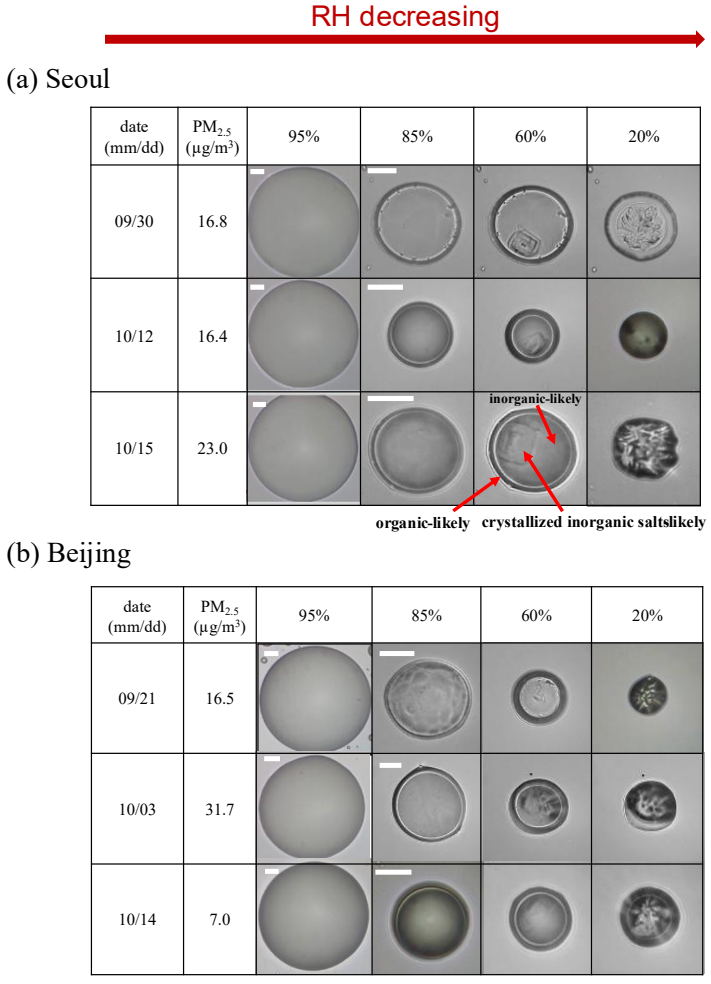

Figure 1. Optical images obtained during RH decrease for (a) Seoul and (b) Beijing $PM_{2.5}$ droplets showing phase separation, as RH decreases from ~95% to 85, 60, and 20 %. Upon dehydration, particles transition from a homogeneous single phase to a core-shell morphology, illustrating separation of organic and inorganic components driven by water loss. The scale bar represents 20 μm. Seoul (10/15) and Beijing (10/14) samples have already been reported by Song et al. (2025) and are included here for completeness of discussion.

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

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

Figure 2 shows the viscosities of $PM_{2.5}$ droplets collected from Seoul and Beijing within the experimentally accessible RH range (RH < ~40%). Across the RH range between ~40 and 25%, the mean viscosities were approximately 10^4–10^8 Pa·s, corresponding to consistencies ranging from peanut butter to tar pitch (Koop et al., 2011; Reid et al., 2018). Within the viscosity framework, the autumn-time urban $PM_{2.5}$ droplets examined here predominantly fell within the semisolid regime. The derived viscosities exhibited substantial sample-to-sample and droplet-to-droplet variability, reflecting compositional heterogeneity and experimental uncertainty. The overall uncertainty of individual viscosity estimates was within approximately two orders of magnitude, primarily driven by assumptions regarding slip length in the fluid-dynamics simulations, as reported previously (Renbaum-Wolff et al., 2013b; Grayson et al., 2015; Grayson et al., 2016).

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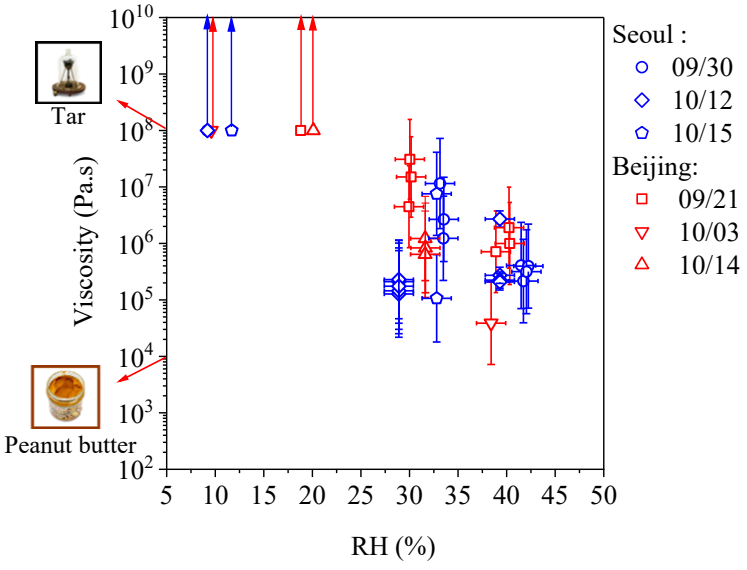

224

225 **Figure 2. Mean viscosities of $PM_{2.5}$ droplets derived from experimentally measured flow times (Fig. S1) using poke-**
226 **and-flow measurements and COMSOL simulations. Markers denote mean values, with *y*-error bars indicating upper**
227 **and lower bound deviations from the mean, calculated as the difference between the mean and the corresponding upper**
228 **and lower bounds derived from simulations using minimum and maximum input parameters (Table S2). Upward**
229 **arrows indicate lower-limit viscosities where no restorative flow was observed within the experimental timescale. The**
230 ***x*-axis error bars represent the RH sensor uncertainty (±1.5%), as determined from our RH sensor calibration in the**
231 **flow cell. Reference viscosities for peanut butter (~10^4 Pa·s) and tar (~10^8 Pa·s) are shown for comparison (Koop et al.,**
232 **2011; Reid et al., 2018).**

233 At lower RH, $PM_{2.5}$ droplets frequently exhibited brittle cracking without observable relaxation (Fig. 4).
234 When no restorative flow was detected over observation periods exceeding two hours, a conservative lower-limit
235 viscosity of ~10^8 Pa·s was assigned. Under these conditions, $PM_{2.5}$ droplets from Seoul exhibited cracking at RH
236 values of ~9.2%, ~9.2%, and ~11.7%, whereas droplets from Beijing cracked at comparatively higher and more

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variable RH values of ~18.8%, ~9.6%, and ~20.1% on each date. These observations indicate that the RH threshold for the transition to non-flowing behavior is dependent on $PM_{2.5}$ composition. The inferred lower-bound viscosities correspond to consistencies comparable to, or exceeding, those of tar-like materials, suggesting extremely viscous properties or arrested internal flow under dry conditions.

(a) Seoul

date (mm/dd)	$PM_{2.5}$ (μg/m³)	RH	pre-poking	poking	after poking	post poking
09/30	16.8	~ 40%			$T = 0$	$\tau_{exp,flow}$ = 97 s
10/12	16.4	~ 40%			$T = 0$	$\tau_{exp,flow}$ = 13 s
10/15	23.0	~ 33%			$T = 0$	$\tau_{exp,flow}$ = 48 s

(b) Beijing

date (mm/dd)	$PM_{2.5}$ (μg/m³)	RH	pre-poking	poking	after poking	post poking
09/21	16.5	~ 40%			$T = 0$	$\tau_{exp,flow}$ = 251s
10/03	31.7	~ 40%			$T = 0$	$\tau_{exp,flow}$ = 15s
10/14	7.0	~ 31%			$T = 0$	$\tau_{exp,flow}$ = 148s

Figure 3. Poke-and-flow results for (a) Seoul and (b) Beijing $PM_{2.5}$ droplets at ~40 – ~30 % RH conditions under which particle flow was observed. The post poking frame corresponds to the $\tau_{(exp,\ flow)}$, defined as the time required for the inner hole diameter to decrease to 50%. The scale bar represents 20 μm.

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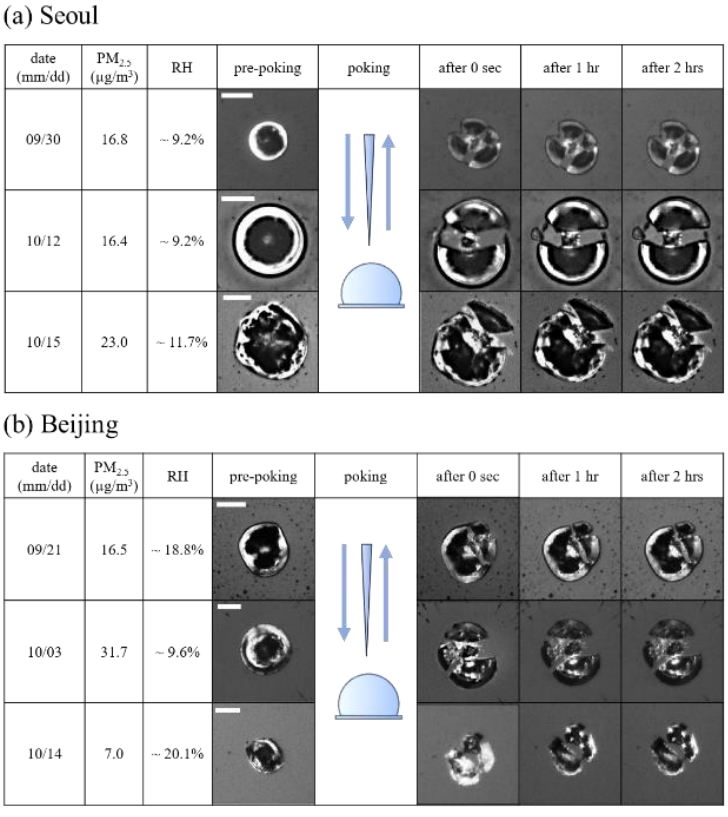

Figure 4. Optical images of (a) Seoul and (b) Beijing $PM_{2.5}$ droplets exhibiting cracking at their respective RH after poking. The scale bar represents 20 μm.

The autumn $PM_{2.5}$ samples examined here were overall organic-rich, with sulfate as the dominant inorganic ion, primarily present as AS (Sect. 3.1). The RH-resolved viscosities measured for $PM_{2.5}$ were compared with laboratory measurements for internally mixed organic–AS systems, commonly used as surrogates for organic-rich, sulfate-containing aerosols (Fig. 5). Previous laboratory studies indicated that citric acid (CA)–AS–H_2O systems exhibited relatively low viscosities and weak RH dependence at comparable RH, whereas sucrose–AS–H_2O systems showed a pronounced increase in viscosity upon dehydration, reaching ~10^3–10^5 Pa·s at RH of ~20–30% and approaching ~10^8 Pa·s upon cracking at lower RH (e.g., sucrose–AS–H_2O for 4:1 and 1:1, Fig. 5) (Jeong

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et al., 2022; Tong et al., 2022; Sheldon et al., 2023). Accordingly, sucrose–AS systems represent the highest RH-dependent viscosities reported among commonly used laboratory surrogates, providing an appropriate upper reference for comparison with organic-rich urban $PM_{2.5}$.

Within the experimentally accessible RH range (≤ ~40%) in this study using the poke-and-flow technique, the viscosities determined for organic-rich urban $PM_{2.5}$ droplets were comparable to the highest values reported for sucrose–AS laboratory systems and, in several cases, exceeded this upper range, while remaining consistently higher than those reported for CA–AS systems at comparable RH. These results demonstrated that organic-rich urban $PM_{2.5}$ attained viscosities at the upper end of RH-dependent viscosity values previously reported for laboratory-generated sucrose–AS–H_2O systems, without extrapolation beyond the measured RH range.

Our conclusions are based on a limited number of filters and droplets; broader seasonal and regional generalizations will require more extensive sampling. In addition, because the experiments were conducted on micrometer-sized extracted droplets on a substrate, future studies should combine rheological measurements with phase-resolved chemical imaging and explicitly examine temperature dependence over a wider, atmospherically relevant range to better constrain viscosity variability under real boundary-layer conditions.

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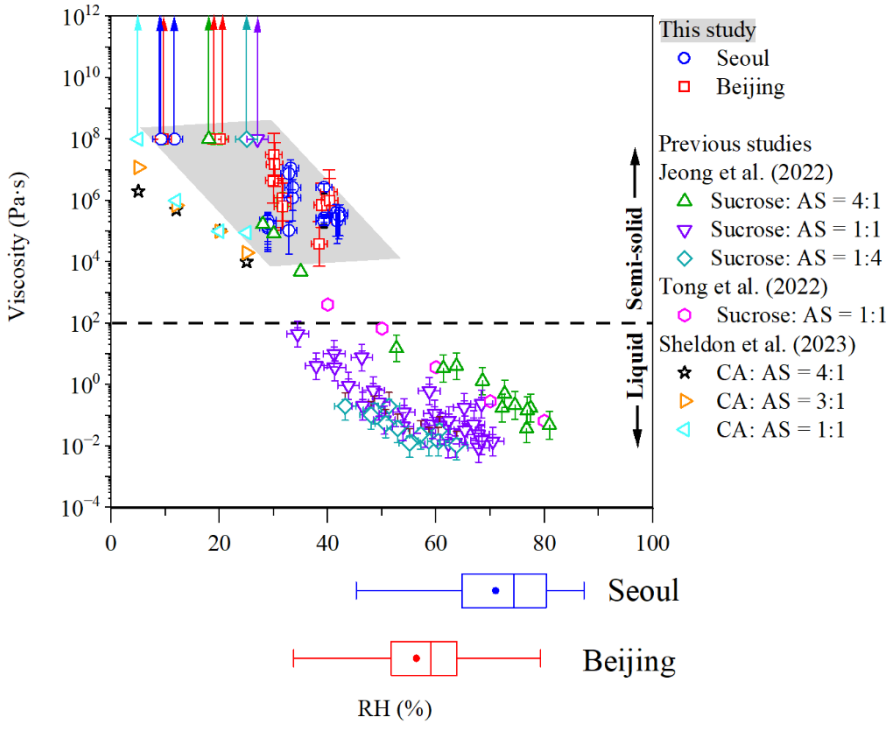

Figure 5. RH-dependent viscosities of Beijing and Seoul $PM_{2.5}$ droplets compared with sucrose–AS–H_2O and citric acid (CA)–AS–H_2O systems from previous studies (Jeong et al., 2022; Tong et al., 2022; Sheldon et al., 2023). *y*-error bars in this study indicate uncertainty ranges of the modeled viscosities for data corresponding to Fig. 2. Box plots in the lower panel show hourly RH distributions in Seoul and Beijing derived from the days on which $PM_{2.5}$ samples were collected, with markers indicating mean values, boxes representing the 25th–75th percentiles, and whiskers showing minimum and maximum values.

4. Conclusion

In this study, we investigated the phase behavior and viscosity of organic-rich urban $PM_{2.5}$ collected during autumn from Seoul and Beijing, using filter extracts. Optical microscopy observations revealed RH-driven phase transitions, including well-mixed single liquid, two liquid phases, three phases, and the development of non-flowing morphologies during dehydration. Using the poke-and-flow technique coupled with fluid-dynamic

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simulations, we quantitatively constrained aerosol viscosity at ~290 K within the experimentally accessible RH range (RH < ~40%). For ~40–25% RH, the inferred viscosities of organic-rich $PM_{2.5}$ spanned approximately 10^4–10^8 Pa·s, corresponding to semisolid to non-flowing behavior on the experimental timescale. At lower RH, brittle cracking without observable relaxation was observed, and conservative lower-limit viscosities of ~10^8 Pa·s were assigned.

When placed in the context of existing laboratory studies, the viscosities inferred for organic-rich urban $PM_{2.5}$ were comparable to the highest values reported for sucrose–AS–H_2O laboratory systems and, in several cases, exceeded this upper range within the RH interval of 20–40%, while remaining consistently higher than those reported for CA–AS systems at similar RH. These findings demonstrated that organic-rich urban $PM_{2.5}$ can attain viscosities at the upper end of RH-dependent viscosity values previously reported for laboratory-generated organic–inorganic aerosol surrogates, without reliance on extrapolation beyond the measured RH range.

Although these viscosity constraints were derived from micrometer-sized droplets prepared from $PM_{2.5}$ filter extracts and therefore do not fully preserve the native morphology or size of ambient submicron particles, the results provide direct, field-based quantitative benchmarks for aerosol viscosity under atmospherically relevant low-RH conditions. Such highly viscous or effectively non-flowing states are expected to slow intra-particle diffusion and internal mixing, with implications for gas–particle partitioning and multiphase chemical processing in organic-rich urban aerosols. Future studies extending these measurements across additional seasons, compositions, and particle sizes will be essential for further constraining the role of aerosol viscosity in urban atmospheric processes.

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Author contributions

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

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Notes

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