Supplement of Atmos. Meas. Tech., 17, 6073–6084, 2024
https://doi.org/10.5194/amt-17-6073-2024-supplement

Atmospheric Measurement Techniques
Open Access

Supplement of

Deriving the hygroscopicity of ambient particles using low-cost optical particle counters

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Aerosol hygroscopicity derived using OPC and BAM data

The optical particle counter (OPC) (model: OPC-N2, Alphasense) provides digital outputs of PM_1, $PM_{2.5}$, PM_{10}, and optionally $PM_{4.25}$, along with histograms of the particle counts for 16 size bins ranging from 0.38 to 17 μm. This device is designed to monitor ambient aerosol concentrations without any drying system attached to the sampling inlet. In contrast, the Beta Attenuation Mass Monitor (BAM) (model: BAM1020, Met One Instrument) is designed to monitor dry particle mass concentrations of $PM_{2.5}$ and PM_{10}, using a heating device to ensure the sampling relative humidity (RH) remains below 50%. If the RH of the sampled air stream exceeds 50%, the inlet heater activates, reducing the RH to approximately 35% by warming the air stream downstream before reaching the filter tape. If the RH is below 50%, the heater remains inactive, not altering the sampling flow RH. The technical specifications for the OPC and BAM are summarized in Table S4.

To derive aerosol hygroscopicity (κ), the sensitivity coefficient of OPC was evaluated first using the data points at RH ≤ 50%, as described in Section 2.3 of the main content. Depending on the size range, $\alpha_{2.5}$, α_{10}, and $\alpha_{2.5\text{-}10}$ represent the sensitivity coefficient of OPC for $PM_{2.5}$, PM_{10}, and $PM_{2.5\text{-}10}$, respectively. For PM_{10}, a range of hygroscopicity of 0 to 1.2 was applied to Eq. (S1) to obtain $M_{d,derived,10}$ and evaluate the mean absolute percentage error (MAPE) between $M_{d,derived,10}$ and $M_{BAM,10}$. MAPE as a function of the applied hygroscopicity is plotted to determine the κ_{10} range, which has MAPE ≤ 1.1×the lowest MAPE, considering the uncertainty. A similar calculation is applied to $PM_{2.5}$, and $PM_{2.5\text{-}10}$ to derive $\kappa_{2.5}$ and $\kappa_{2.5\text{-}10}$ directly.

$$M_{d,derived,10} = \alpha_{10} \times M_{OPC,10} \times \left[\left(\frac{S\,\kappa_{10}}{1-S}\right) \times \frac{\rho_w}{\rho_d} + 1\right]^{-1} \tag{S1}$$

However, PM_{10}^* in Table 1 considers the size-dependent sensitivity with a mean hygroscopicity. κ_{10} is derived using the following equation:

$$M_{d,derived,10} = \left(\alpha_{2.5} \times M_{OPC,2.5} + \alpha_{2.5-10} \times M_{OPC,2.5-10}\right) \times \left[\left(\frac{S\,\kappa_{10}}{1-S}\right) \times \frac{\rho_w}{\rho_d} + 1\right]^{-1} \tag{S2}$$

The derived κ_{10} using Eq. (S2) ranges from 0.13 to 0.23, falling between κ values for $PM_{2.5}$ and $PM_{2.5\text{-}10}$, and is more reasonable than from Eq. (S1), as discussed in Section 3.2.2.

Table S1: Summary of the applied sensors in AQB and instruments at TW-EPA station.

	Sensors in AQB (Manufacturer)	Detection range, Detection resolution	Instruments at TW-EPA (Manufacturer)	Detection range, Detection resolution
T, RH	SHT31 (Seeed)	T: -40 – 125 ℃, 0.1 ℃ RH: 0 – 100%, 0.1%	083D (Met One Instruments)	T: -30–50 ℃, 0.1 ℃ RH: 0–100%, 0.04%
CO	CO-B4 (Alphasense)	0–1000 ppmv, in ppbv	APMA360 (Horiba)	0–100 ppmv, 0.02 ppmv
NO	NO-B4 (Alphasense)	0–20 ppmv, in ppbv	ML9841 (Horiba)	0–20 ppmv, 1 pptv
NO_2	NO2-B43F (Alphasense)	0–20 ppmv, in ppbv	ML9841 (Horiba)	0–20 ppmv, 1 pptv
O_3	OX-B431 for O_3+NO_2 (Alphasense)	0–20 ppmv, in ppbv	ML9810 (Ecotech)	0–20 ppmv, 1 pptv
SO_2	SO2-B4 (Alphasense)	0–100 ppmv, in ppbv	ML9850 (Ecotech)	0–20 ppmv, 1 pptv
VOC	PID-AH2 (Alphasense)	0–40 ppmv, in ppbv	APHA360 (Horiba)	0–100 ppmv, 0.022 ppmv
PM	OPC-N2 (Alphasense)	0.01–1500 μg m^{-3}, 0.1μg m^{-3}	BAM1020 (Met One Instruments)	0–10,000 μg m^{-3}, 0.1μg m^{-3}

Table S2: The hygroscopicity, molecular weight, and density of salts used in deriving hygroscopicity of mixtures.

salt	$(NH_4)_2SO_4$	$(NH_4)HSO_4$	NH_4NO_3	$NaNO_3$	NaCl
hygroscopicity	0.61[a]	0.80[c]	0.67[a]	0.88[a]	1.28[a]
molecular weight (g mol^{-1})	132	115	80	85	58.5
density (g cm^{-3})	1.70[b]	1.78[b]	1.72[b]	2.26	2.17

[a] hygroscopicity summarized in Petters and Kreidenweis (2007) as CCN derived hygroscopicity.

[b] density of chemical species reported by Xu et al. (2020).

[c] hygroscopicity calculated by Kim et al. (2021)

Table S3: The correlation coefficient (r) for measured parameters between two AQB systems and between AQB no. 1 and the TW-EPA Nanzi station

r	AQB no. 1 vs AQB no. 2	AQB no. 1 vs TW-EPA
T	0.958	0.948
RH	0.949	0.932
CO	0.995	0.976
NO	0.976	0.624
NO_2	0.944	0.504
Ox (NO_2+O_3)	0.979	0.961
VOC	0.675	-0.373
SO_2	0.973	0.343
$PM_{2.5}$	0.978	0.689
PM_{10}	0.967	0.483

Table S4: Technical specifications for OPC and BAM

	OPC	BAM
Manufacturer	Alphasense	Met One Instruments
Model	OPC-N2	BAM1020
Particle range (µm)	0.38 – 17	-
Bin number	16	-
Laser wavelength (nm)	658	-
Refractive index	1.5 + 0i	-
Setting particle density (g cm^{-3})	1.65	-
Max particle count rate (s^{-1})	10,000	-
Detection range (µg m^{-3})	0.01-1500 (for PM_{10})	0 – 10,000
Measurement resolution (µg m^{-3})	0.1	0.1
Sampling flow rate (LPM)	~5	16.67
Coincidence probability (% at 10^6 L^{-1})	0.84	-
Unit dimensions (L × D × H)	75 × 60 × 63.5 (mm)	36.2 × 43.2 × 46.7 (cm)
Weight	105 g	19 kg
Particle size designations (µm)	-	PM_{10}, $PM_{2.5}$, and $PM_{2.5-10}$
Filter tape	-	Continuous glass fiber filter

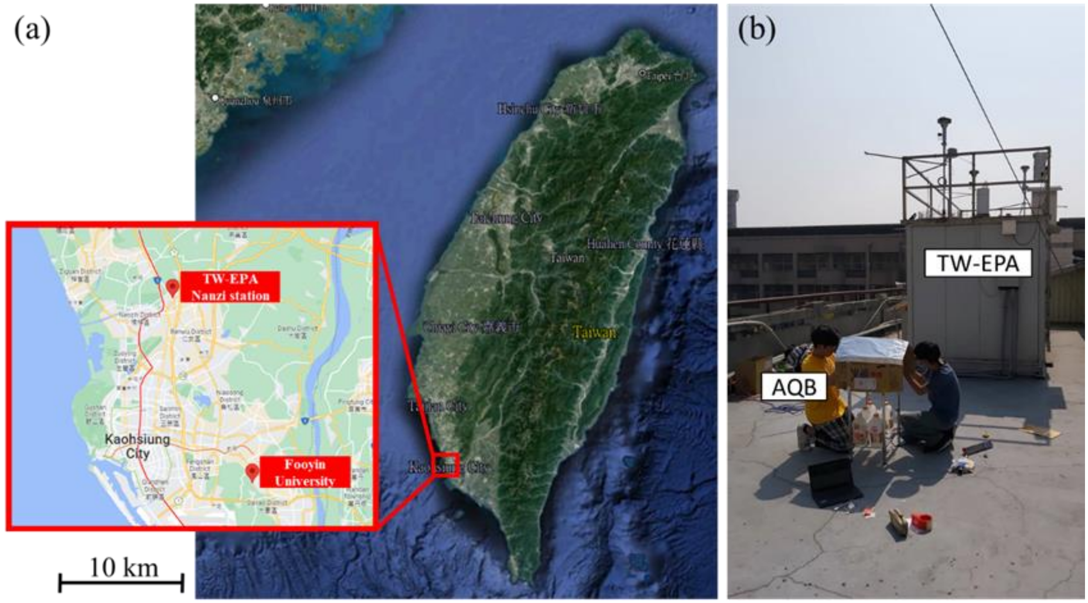

Figure S1. (a) Location of TW-EPA Nanzi station (AQB calibration campaign) site and Fooyin University (2013 sampling campaign). (from © Google Earth 2024 and © Google Maps 2024). (b) Photograph of the setup in AQB system operational state. AQB was located approximately 5 m horizontally and 2 m vertically from the sampling inlet of TW-EPA Nanzi station).

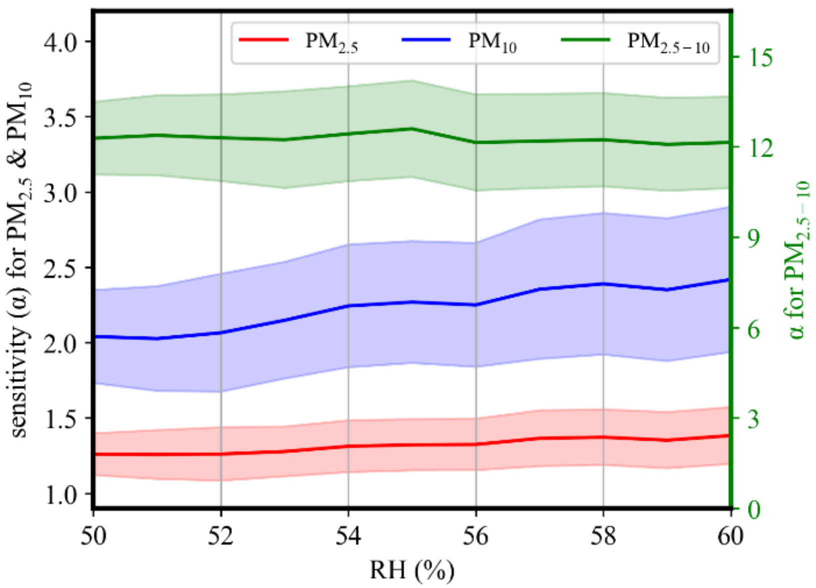

Figure S2: The determined sensitivity as a function of RH thresholds for $PM_{2.5}$ (red), PM_{10} (blue) and $PM_{2.5-10}$ (green). The shaded area represents the mean value ± 0.5σ

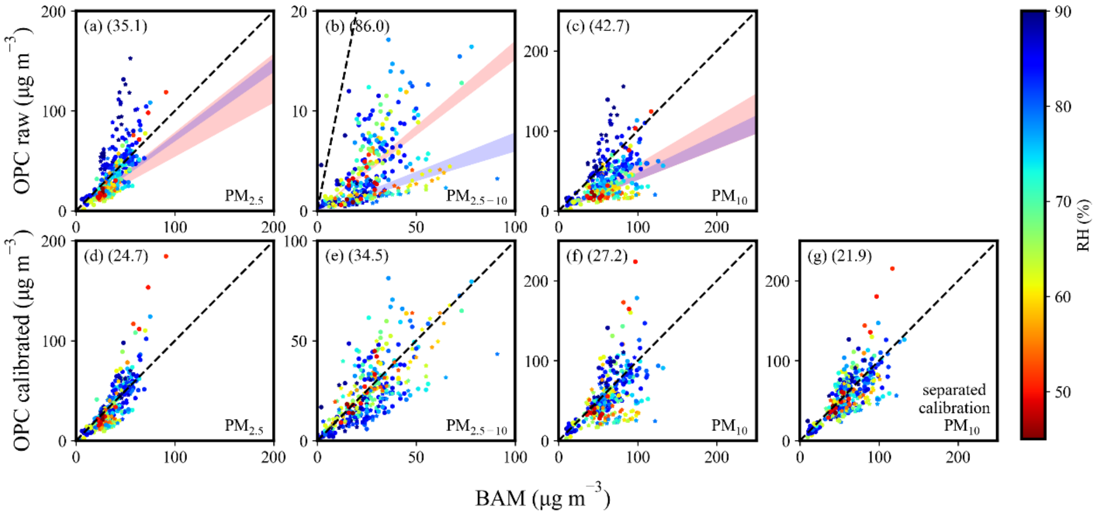

Figure S3: The correlation of mass concentration between BAM and OPC in AQB no. 2: (a, d) $PM_{2.5}$, (b, e) $PM_{2.5\text{-}10}$, (c, f) PM_{10}, and (g) separated calibration PM_{10}. (a-c) are the raw data, while (d-g) are the calibrated data. The marker color corresponds to RH. The shaded region represents the data associated with the sensitivity coefficient ("α"). The data show the first period (red paved/circle points) and the second period (purple paved/star points). The value in parentheses is the MAPE in percentage.

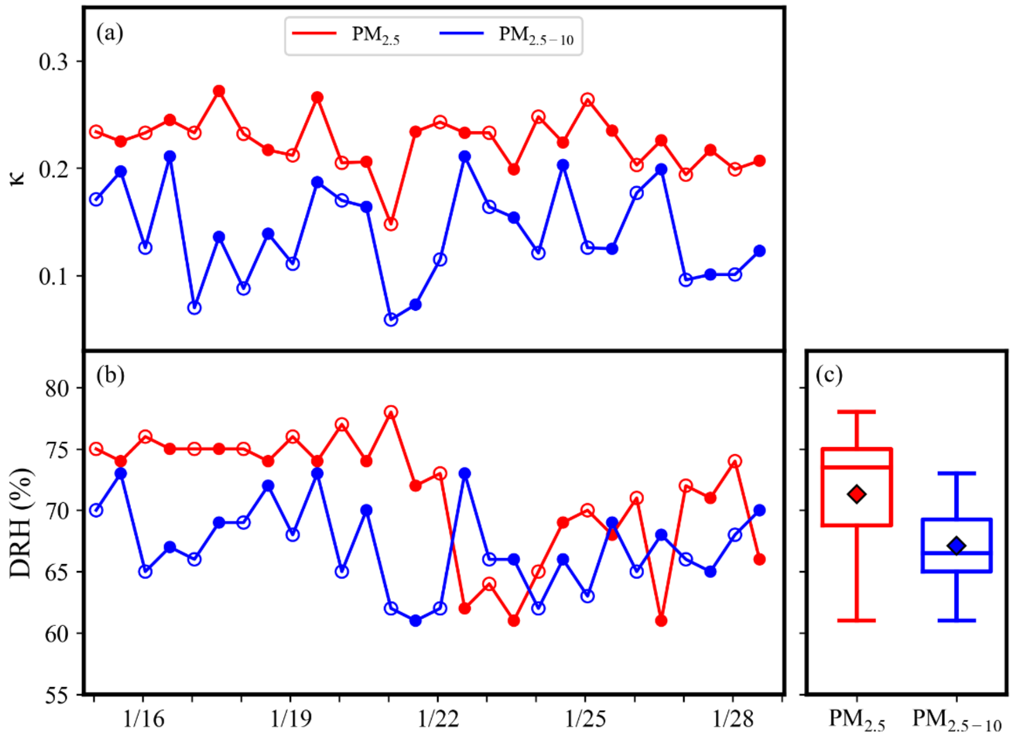

Figure S4: The temporal profiles of (a) derived κ from IC data, (b) DRH determined from E-AIM, and (c) DRH box-plot distribution for the 2013 winter campaign period. (hollow circle: daytime samples; solid circle: nighttime samples; diamond: mean value; outliers: < 1st quartile Q1-1.5 interquartile range (IQR) or > 3rd quartile Q3+1.5 IQR).

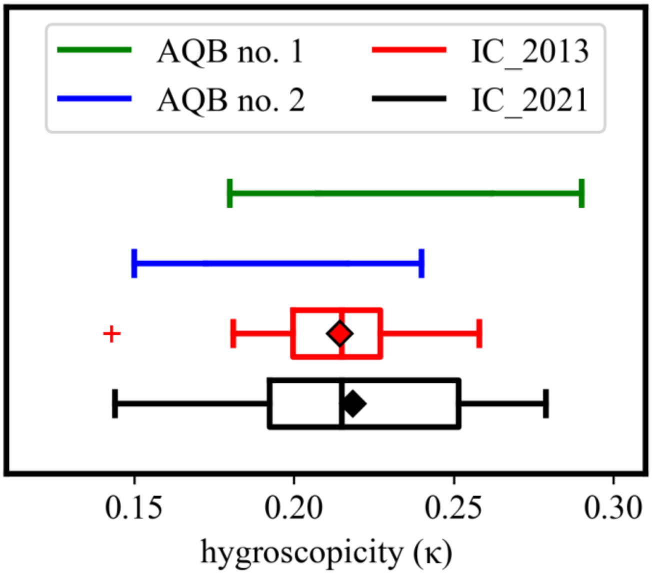

Figure S5: The hygroscopicity of $PM_{2.5}$ derived from OPC and IC data with an assumed particle density of 1.2 g cm^{-3}. The IC_2021 is from 2021 samples collected at the National Kaohsiung University of Science and Technology (22°46'22.4" N 120°24'03.4" E) in Kaohsiung for the period of 8 – 18 December 2021. (diamond: mean value; outliers: < 1st quartile Q1-1.5 interquartile range (IQR) or > 3rd quartile Q3+1.5 IQR).

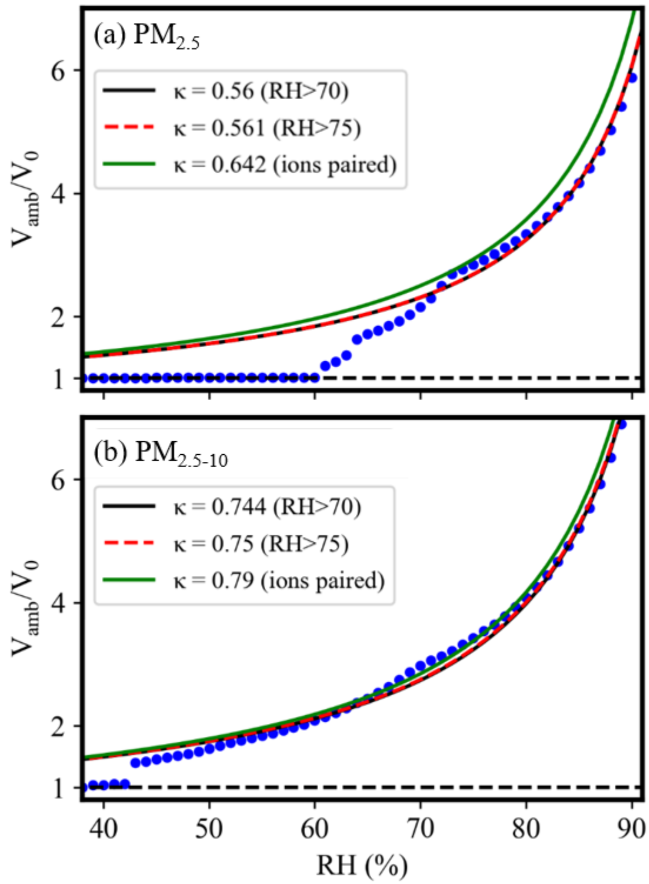

Figure S6: The volume ratio (ambient state compared to dry state) as a function of RH for (a) $PM_{2.5}$ and (b) $PM_{2.5\text{-}10}$ using E-AIM, along with the fitting lines using κ-Köhler equation (Eq. 2) with data points above the threshold as indicated in the legend. Sample mean composition with the molarity ratio of Na^+:NH_4^+:Cl^-:SO_4^{2-}:NO_3^- is 7:229:0:71:94 and 65:59:16:19:70 for $PM_{2.5}$ and $PM_{2.5\text{-}10}$, respectively. No insoluble composition is taken into account in the calculation.

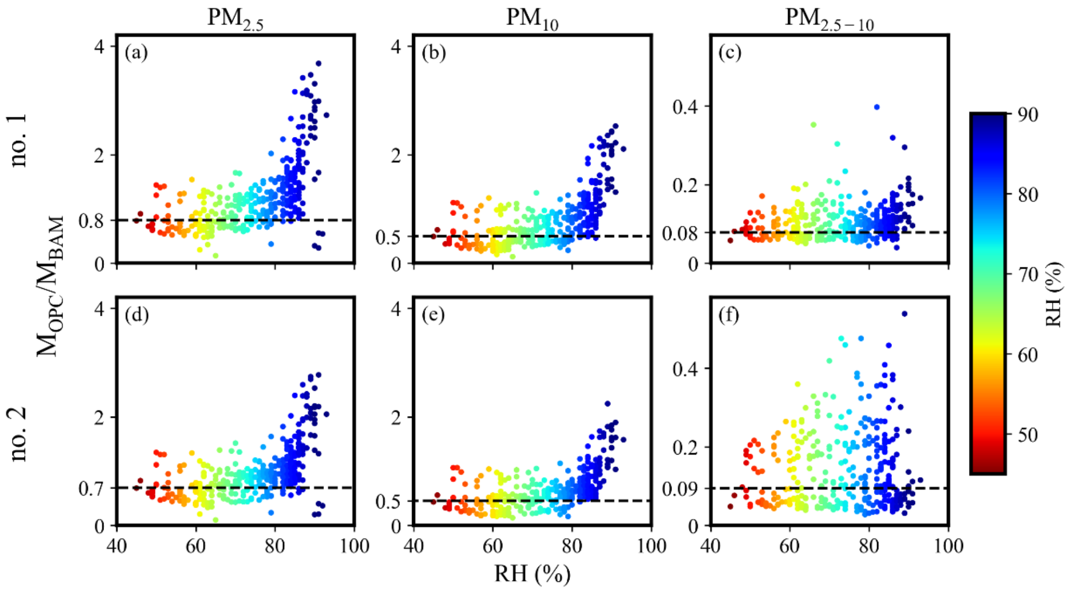

Figure S7: The mass ratio (ambient state compared to dry state) as a function of RH for (a, d) $PM_{2.5}$, (b, e) PM_{10}, and (c, f) $PM_{2.5\text{-}10}$ for AQB #1 and #2 data comparison. The marker color corresponds to RH. The dashed lines indicate the inverse of the sensitivity coefficient (α) obtained from data at RH ≤50%.

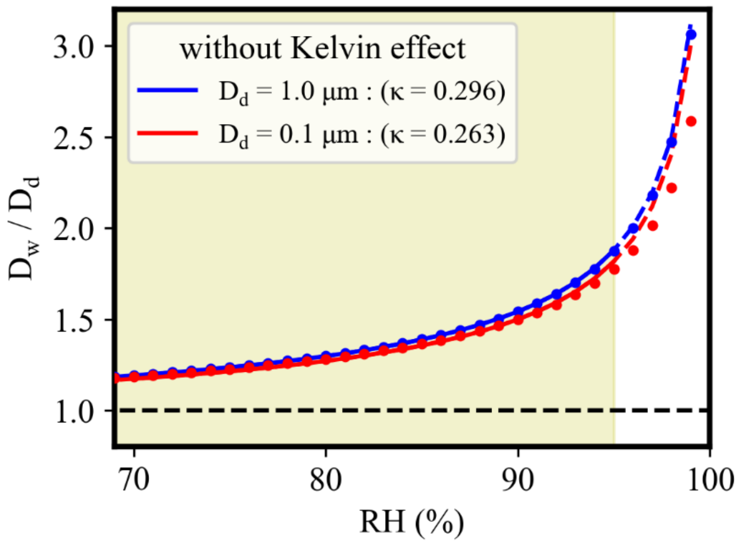

Figure S8: The particle growth diameter ratio as a function of RH for particle sizes of 0.1 µm (red) and 1.0 µm (blue). Points are diameter ratio with the Kelvin effect considered at κ = 0.3 for 70-95% of RH using Eq. 2, and solid lines are the fitting results for the points to derive κ without the Kelvin effect term.

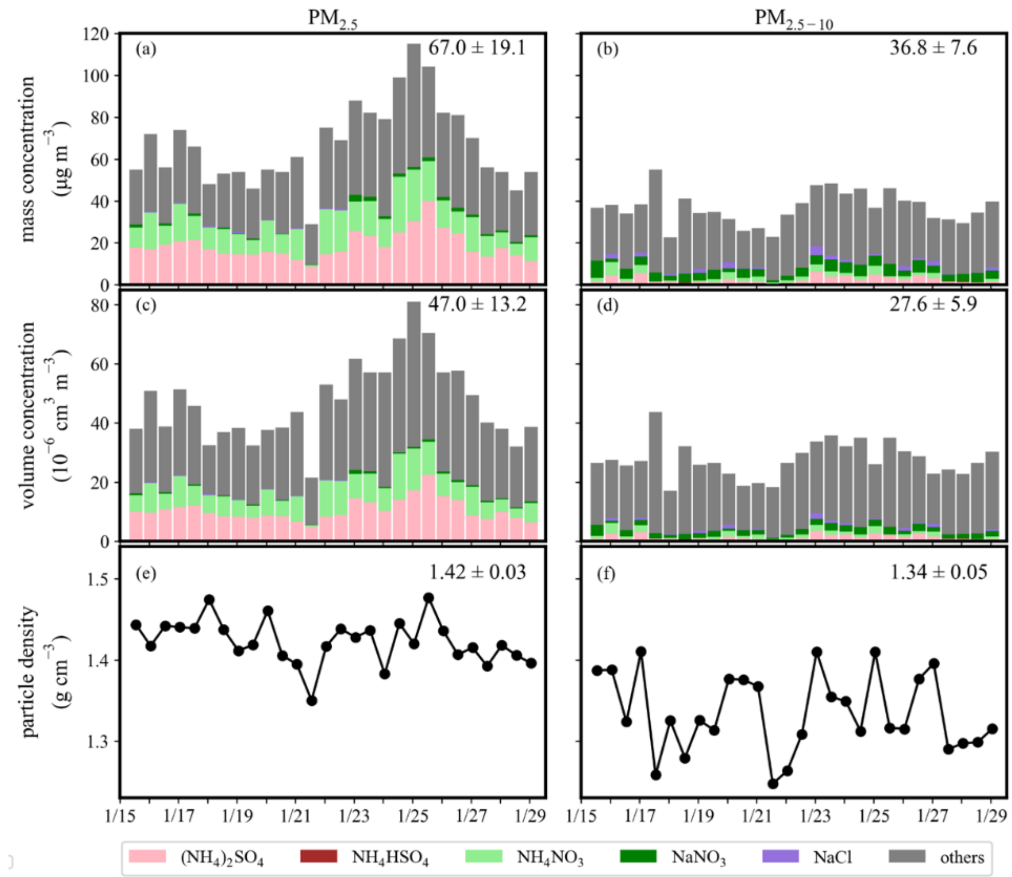

Figure S9: The temporal profiles of (a,b) mass and (c,d) volume concentration of chemical species from IC analysis and (e,f) calculated particle density for $PM_{2.5}$ and $PM_{2.5\text{-}10}$. The column color corresponds to the contribution of different components, with others characterized as secondary organic compositions having a density of 1.2 g cm^{-3}. The number on the upper right corner is the mean ± 1 SD.

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