

Complex hygroscopic behaviour of ambient aerosol particles revealed by a piezoelectric technique

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List of supporting materials:

1. Seven texts, Text S1 - Text S7
2. Two tables, Table S1 – Table S2
3. Five figures, Figure S1 – Figure S5
4. References

Text S1. Sampling Site

The sampling was conducted at the Natural Aerosol and Bioaerosol High Altitude (NABHA) Laboratory at the College of Engineering, Munnar (10.0930° N, 77.0682° E), located in the Western Ghats of India, one of the most significant biodiversity hotspots in the world (Kale et al., 2016). The site is at about ~1600 m in elevation and 90 km from the Arabian Sea. Munnar possesses a moderately rugged topography with high mountain peaks and deep river valleys covered with diversified vegetated areas, tea plantations, barren land/rocky areas, and water bodies (Gupta et al., 2020). During the southwest monsoon season (June-September), the Western Ghats block the south-westerly winds, causing rainfall on these mountain ranges.

Text S2. Instrumentation

The ambient aerosols were collected on polytetrafluoroethylene (PTFE) membrane filters (pore size 0.2 μm) using a Micro-Orifice Uniform Deposit Impactor (MOUDI) with a sampling flow rate of 30 L/min. The MOUDI-II 120R (TSI) utilized 10 rotating stages to achieve a uniform deposit of particles on the filters, with nominal cut-off points at 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, and 10 μm (Marple et al., 2014). Concurrently, various meteorological parameters such as temperature, humidity, pressure, wind speed, wind direction, and precipitation were recorded using an Automatic Weather Station (AWS, Clima Sensor US) at the sampling location throughout the sampling period.

The number size distribution of the ambient aerosol particles was measured using a Scanning Mobility Particle Sizer (SMPS), which comprises a 3082 Electrostatic Classifier with 3081 Long Differential Mobility Analyzer (L-DMA, TSI Model 3081) and a butanol-Condensation Particle Counter (CPC, TSI Model 3750). The measurements were carried out with an aerosol flow rate of 1 L/min, spanning a size range of 10.2 – 430 nm. The aerosol mass distribution measurements for the larger-sized particles (0.3 – 20.0 μm) obtained using an Ultraviolet Aerodynamic Particle Sizer (UV-APS, TSI Inc., Model 3314) from a campaign conducted at

the same location during the Monsoon season of 2014 (June-August) were also incorporated in the study.

The hygroscopicity of aerosol particles was analyzed using a sensitive mass-balance Quartz Crystal Microbalance (QCM, QSense Analyzer, Biolin Scientific), which can measure the mass changes at the nanograms level. In 1959, Sauerbrey proposed that an adsorbed mass (Δm) causes a decrease in the resonant frequency (Δf) of the quartz crystal, and their linear relationship is given by,

$$\Delta f = -\frac{C}{n}\Delta m \quad (\text{S1})$$

where C ($=17.7 \text{ ng}\cdot\text{cm}^{-2}\cdot\text{Hz}^{-1}$ for 5 MHz AT-cut crystals) is the mass sensitivity constant specific to the quartz resonator and n ($=1,3,5,\dots$) is the overtone number (Sauerbrey, 1959). The Sauerbrey equation is valid only when the film deposited on the QCM quartz sensor is rigid and perfectly coupled to the sensor surface (Rodahl & Kasemo, 1995). QCM experiments were performed with a 5 MHz AT-cut quartz sensor coated with SiO_2 , and the particles were uniformly deposited on the sensor. The ambient aerosol particles collected on PTFE filters were directly transferred to the QCM sensor by placing the filter on the sensor and then gently pressing it using a cotton piece. The particles on the sensor were examined under the microscope to ensure the uniformity of the film.

The relative humidity (RH) was incrementally increased from 2% to 93% to promote the uptake of water by aerosol particles, resulting in a corresponding increase in mass. The QCM provides the fundamental frequencies corresponding to the mass deposited on the sensor. Measurements were taken at various RH points in the sub-saturated region for a blank sensor and a sensor coated with aerosol particles. The difference in the frequencies between the coated and the blank sensor at a particular RH was subsequently converted to the mass of water absorbed or adsorbed at that point using the Sauerbrey equation.

The effective particle hygroscopicity, κ_{CCN} (Petters & Kreidenweis, 2007) under supersaturated conditions was determined using size-resolved Cloud Condensation Nuclei (CCN) measurements acquired through a CCN counter (CCNC, CCN-100, DMT), employing the method outlined in Rose et al. (2008).

Text S3. Experimental setup

The schematic experimental setup designed to measure the hygroscopicity of the aerosol particles using QCM is depicted in Figure S1. Dry nitrogen was passed through a HEPA filter (Whatman 6702-9500 HEPA-CAP filters) and divided into two channels, one of which was humidified using a water bubbler. Desired RH values were created by controlling the flow ratio of dry and humidified air from both channels using digital mass flow controllers (MFCs) (Alicat Scientific–50 SCCM). The RH was continuously monitored at various points in the flow pathway using thermohydrometers (testo 605i). The accuracy of the RH values in the ranges 10-35%, 35-65%, 65-90% and <10% or >90% are $\pm 3.0\%$, $\pm 2.0\%$, $\pm 3.0\%$ and $\pm 5\%$, respectively. Each humidity condition was sustained until an equilibrium condition was reached, where no further evaporation and condensation happened. The amount of water absorbed by the particles at different values of RH in the sub-saturated region was measured continuously by the QCM. The QCM module temperature was kept at least 0.5 °C higher than the room temperature to avoid condensation of water vapour onto the particles inside the QCM module. The experimental setup employed a low flow range of 0-50 sccm, and any variations in the flow could significantly affect the experimental results. To mitigate this issue, the flow rate was checked at various control points before and after each experiment using a Gilibrator (Sensidyne).

To ensure accurate measurements, it is crucial to properly clean the sensor surface before depositing the particles. The sensors were cleaned with milli-Q water and methanol, then dried with N₂ gas. Subsequently, a UV-Ozone treatment was applied to eliminate various contaminants, particularly organic compounds, from the SiO₂-coated sensor. The treatment was done for 20 minutes at a power of 1 mW/cm² immediately before depositing the sample particles on the sensor.

Text S4. Mass-based hygroscopic growth factor (gf_m) for laboratory-generated particles

The QCM technique for determining hygroscopicity was validated for a sucrose thin film. The mass-based hygroscopic growth factor (gf_m , mass ratio of the film at an elevated RH to that at <5% RH) for sucrose particles for a range of RH (2-93%) was calculated from QCM measurements, and the results were compared with the models for testing the accuracy of the method (Liu et al., 2018).

The steps followed for making a sucrose thin film on the sensor are as follows.

1. Prepared different concentrations (0.05%, 0.15, 0.25%, 0.5% etc) of sucrose solution by dissolving sucrose in milli-Q water.

2. Cleaned the sensor surface using Milli-Q water and methanol and treated with UV-Ozone for 20 minutes.
3. The desired volume of the solution was pipetted onto the sensor surface, which was placed on a clean petriplate.
4. The sensor was then placed inside a muffle furnace with a preset temperature of 80°C for 20 minutes to remove water.
5. The sensor with the film was viewed under a microscope (Dino-Lite Edge) to analyze the uniformity of the film.
6. The sensor with a thin film of sucrose was then mounted inside one of the modules of the QCM analyzer.
7. Dry air was allowed to pass over the film for 10-12 hours to further remove the moisture content in the particles.

After checking the uniformity of the film and the stability of different overtones using QSoft software (Fig. S2), the experiment was started with a preset program in FlowVision (software that helps to control the flow rates at the MFCs). The flow rates were changed accordingly in the two MFCs for creating different humidity values starting from dry conditions (<5% RH) to a maximum of 93% RH.

The hygroscopic growth factor values obtained from the QCM measurements were in good agreement with the literature (Norrish, 1966; Starzak & Peacock, 1997; Zobrist et al., 2011) (Fig. S3).

Text S5. Mass-based hygroscopicity parameter (κ_m) for ambient aerosol particles

After transferring the aerosol particles from the filters to the sensor, the sensor was mounted inside the QCM module and the RH was incrementally increased to investigate the water uptake behavior of particles under varying humidity conditions.

We calculated the mass-based hygroscopicity parameter, κ_m , which measures the aerosol particles' ability to take up atmospheric moisture, from the growth factor values obtained from the QCM results (Petters & Kreidenweis, 2007).

$$\kappa_m = \left(\frac{1}{a_w} - 1\right)(gf_m - 1) \quad (\text{S2})$$

where a_w is the water activity and gf_m is the mass-based growth factor of the particle at different RH values. The a_w was approximated as the fractional RH under equilibrium conditions. κ_m could be converted to a volume-based hygroscopicity parameter κ_v , using the following relationship.

$$\kappa_v = \kappa_m \times \frac{\rho_d}{\rho_w} \quad (\text{S4})$$

where ρ_d is the effective density of the dry particle and ρ_w is the density of water (Mikhailov et al., 2013).

The experiment was repeated for the aerosol particles from different stages of MOUDI to carry out a size-resolved study. In this study, the ambient particles were solid and rigid until they deliquesce at higher RH conditions. The QCM can measure the mass of the particles under different RH conditions till they reach the phase transition point (deliquescence relative humidity, DRH).

Text S6. Estimation of Deliquescence Relative Humidity (DRH) and Aerosol Liquid Water Content (ALWC)

The phase transition behavior of ambient aerosols was investigated by analyzing the QCM frequency response curves. During the humidification cycle, the DRH point is identified as the RH at which the oscillation frequencies of QCM response start increasing upon the addition of water (Arenas et al., 2012; Chao et al., 2020). A percentage value, Δf_N , is obtained by normalizing the oscillation frequency change induced by the aerosol particles at a given RH (Δf_R) to the oscillation frequency change resulting from a dry sample (Δf_S at RH <5%) (Chao et al., 2020).

$$\Delta f_N = \left(\frac{\Delta f_R}{\Delta f_S} \right) \times (-100)\% \quad (\text{S4})$$

The negative sign indicates that the oscillation frequency of the sensor decreases when RH increases due to water uptake by the aerosol particles. Δf_N and its derivative with respect to the measured RH, $d(\Delta f_N)/d(\text{RH})$ were plotted for different size ranges of ambient aerosol samples (Fig. 3). The deliquescence of the particles is indicated by an increase in Δf_N or a positive value of $d(\Delta f_N)/d(\text{RH})$. The RH at which $d(\Delta f_N)/d(\text{RH}) \geq 1$ is considered as the DRH point for the respective sample (Chao et al., 2020). Following this method, the DRH for $(\text{NH}_4)_2\text{SO}_4$ particles was estimated between 76-80% from the QCM measurements (Fig. S4), which agrees with the literature values (Arenas et al., 2012; Martin, 2000; Peng et al., 2022).

Similarly, the DRH values for size-resolved ambient aerosol particles from Munnar were estimated.

Aerosol Liquid Water Content (ALWC) at different RH conditions were also estimated from the size-resolved hygroscopic growth factor values combined with the particle number size distribution (PNSD) measurements, assuming volume conservation during the aerosol hygroscopic process (Bian et al., 2014).

$$ALWC = \left[\frac{\pi}{6} \sum_i N_i D_{d,i}^3 (gf(D_d, RH)^3 - 1) \right] \rho_w \quad (S5)$$

where N_i represents the number concentration of dry particles of the i th bin obtained from SMPS and UV-APS measurements, $D_{d,i}$ is the particle diameter, and ρ_w is the density of water.

Text S7. Correction for the RH values inside the QCM module

The flow rate of humidified air in the setup can be impacted by temperature instabilities, potentially affecting the results. To prevent water vapor condensation, it is crucial to maintain the QCM module temperature at a level equal to or higher than the room temperature. To account for any temperature changes inside the QCM module, the RH was corrected using the Clausius-Clapeyron equation.

$$\ln \frac{P_1}{P_2} = \frac{\Delta H_{vap}}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right) \quad (S6)$$

where P_1 and P_2 are the vapor pressures at two temperatures, T_1 and T_2 , ΔH_{vap} is the enthalpy (heat) of vaporization, and R is the gas constant ($8.3145 \text{ J mol}^{-1} \text{ K}^{-1}$). The actual RH value was determined by computing the ratio of actual vapor pressure to saturation vapor pressure, expressed as a percentage.

Table S1. Mass-based hygroscopic growth factor (g_{f_m}) and the corresponding hygroscopicity parameter (κ_m) at different RH values in the subsaturated region for different size ranges of ambient aerosol particles ($<10 \mu\text{m}$) sampled from Munnar using MOUDI. The measurements were obtained from the QCM experiments.

Particle size range (μm)											
<0.056				0.056-0.1				0.1-0.18			
RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v
2.87	1.000			5.02	1.000			3.45	1.000		
11.47	1.004	0.033	0.040	16.90	1.011	0.054	0.064	16.86	1.018	0.088	0.105
27.93	1.009	0.008	0.010	31.96	1.026	0.055	0.065	31.77	1.025	0.054	0.064
43.74	1.015	0.020	0.024	44.03	1.054	0.069	0.082	44.37	1.050	0.062	0.075
58.31	1.020	0.014	0.017	54.65	1.089	0.074	0.088	54.88	1.068	0.056	0.067
66.73	1.027	0.013	0.016	64.93	1.145	0.078	0.094	66.04	1.098	0.051	0.061
72.97	1.034	0.013	0.015	73.29	1.282	0.103	0.124	75.49	1.189	0.062	0.074
80.74	1.051	0.012	0.015	79.76	1.426	0.108	0.130	90.15	1.722	0.079	0.095
				86.56	1.549	0.085	0.102	86.85	1.567	0.086	0.103

Particle size range (μm)											
0.18-0.32				0.32-0.56				0.56-1.0			
RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v
2.95	1.000			3.47	1.000			3.09	1.000		
12.42	1.008	0.059	0.071	12.26	1.027	0.194	0.232	12.34	1.030	0.210	0.251
21.84	1.025	0.090	0.109	22.58	1.055	0.188	0.226	21.50	1.056	0.203	0.244
31.36	1.038	0.083	0.099	30.21	1.079	0.182	0.218	30.57	1.079	0.179	0.214
47.70	1.055	0.060	0.072	47.33	1.158	0.175	0.211	46.91	1.149	0.169	0.203
55.62	1.071	0.057	0.068	55.83	1.232	0.183	0.220	55.25	1.220	0.178	0.214
64.23	1.145	0.081	0.097	64.13	1.365	0.204	0.245	63.93	1.369	0.208	0.250

Particle size range (μm)											
1.0-1.8				1.8-3.2				3.2-5.6			
RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v	RH (%)	g_{f_m}	κ_m	κ_v
3.74	1.000			2.80	1.000			3.33	1.000		
12.14	1.061	0.438	0.526	12.39	1.027	0.191	0.229	11.63	1.013	0.097	0.116
19.58	1.110	0.451	0.541	21.64	1.053	0.191	0.229	19.70	1.029	0.119	0.143
27.80	1.167	0.435	0.522	31.09	1.100	0.223	0.267	28.52	1.053	0.134	0.161
35.00	1.244	0.452	0.543	39.25	1.145	0.225	0.270	35.55	1.056	0.101	0.121
42.85	1.315	0.421	0.505	47.91	1.180	0.195	0.234	43.24	1.075	0.099	0.119
47.37	1.411	0.457	0.548					50.94	1.123	0.119	0.142
								59.05	1.180	0.125	0.150

Particle size range (μm)			
5.6-10.0			
RH (%)	g_{f_m}	κ_m	κ_v
3.14	1.000		
12.25	1.015	0.107	0.128
21.61	1.030	0.110	0.132
31.22	1.046	0.101	0.121
39.43	1.067	0.103	0.123
48.00	1.131	0.142	0.171
56.41	1.177	0.137	0.164
64.77	1.198	0.108	0.129

Table S2. The average mass concentration ($\mu\text{g m}^{-3}$) and the mass fraction of the chemical species in NR-PM₁ measured by aerosol chemical speciation monitor (ACSM) during the period from 06.06.2021 to 27.07.2021.

NR-PM₁ Species	Mass concentration ($\mu\text{g m}^{-3}$)	Mass fraction
Organics	1.15	0.50
Sulphate	0.79	0.35
Nitrate	0.08	0.04
Ammonium	0.22	0.09
Chloride	0.04	0.02

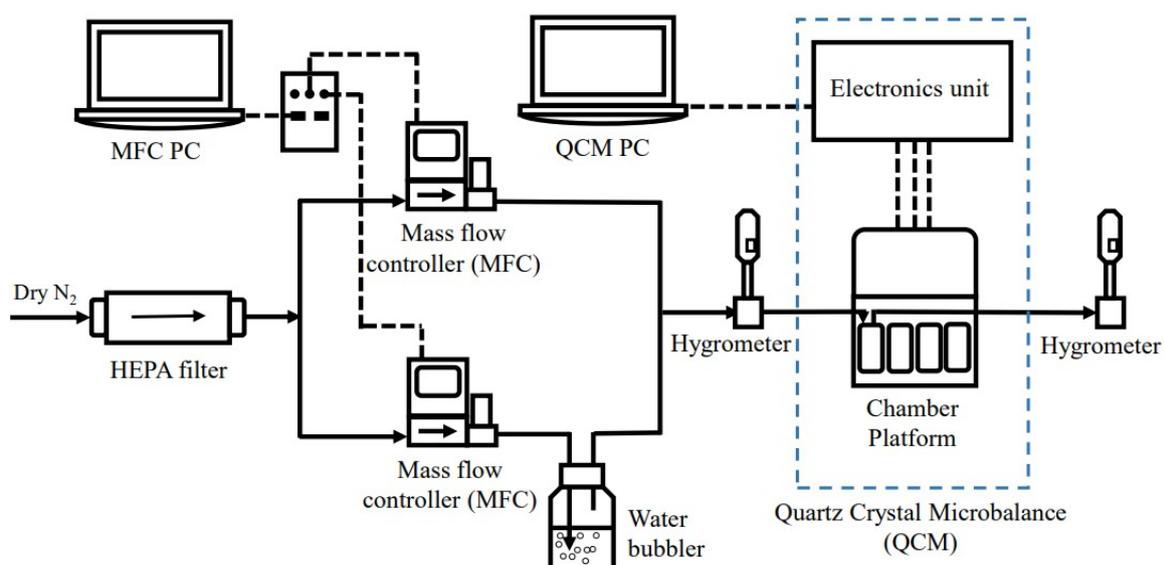


Figure S1. Schematic diagram of the experimental setup for providing a wide range of humidity conditions (2-93%) to study the aerosol-water vapor interactions of the samples coated on the quartz crystal microbalance (QCM) sensor mounted inside the module of the chamber platform. Different humidity conditions were created over the aerosol particles by controlling the flowrates of dry and humidified air using the two mass flow controllers (MFCs) connected in parallel. The RH of incoming and outgoing air is measured using two thermohygrometers connected before and after the QCM chamber platform.

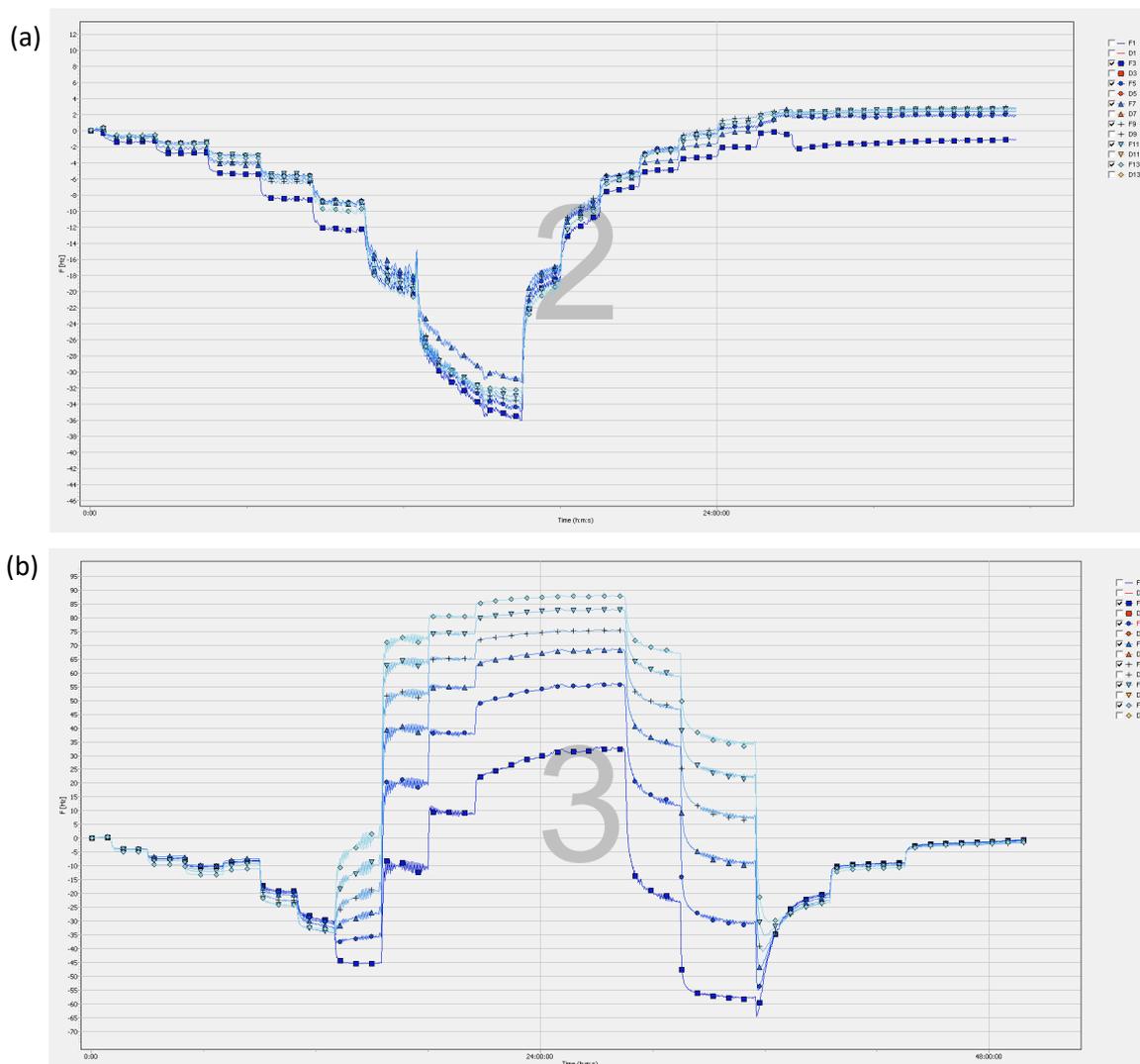


Figure S2. The screenshot of the QSoft software showing the oscillation frequency changes of the quartz crystal microbalance (QCM) sensor with time during the humidifying and drying phase of a QCM experiment for the size ranges 56-100 nm and 560 nm-1 μm of ambient aerosol particles from Munnar. (a) shows a decrease in the oscillation frequency change for the particles in the size range 56-100 nm, corresponding to an increase in mass due to water uptake during the humidifying (increasing RH) phase and a further increase in the frequency change when the water content is removed during the drying (decreasing RH) phase. (b) shows an initial decrease in the frequency change due to water uptake by the aerosol particles in the size range 560 nm-1 μm in the lower RH region (<61.2%) during the humidification cycle. The increase in frequency changes and the separation of overtones at RH=61.2% in the humidifying phase indicate that the particles undergo phase transition (deliquescence) at high RH region and become aqueous droplets leading to less viscous and non-rigid particles on the sensor. The initial frequency change value corresponding to the dry particle mass is obtained after the humidification and drying phase, indicating no particle loss. Different curves correspond to different overtone frequencies of the sensor.

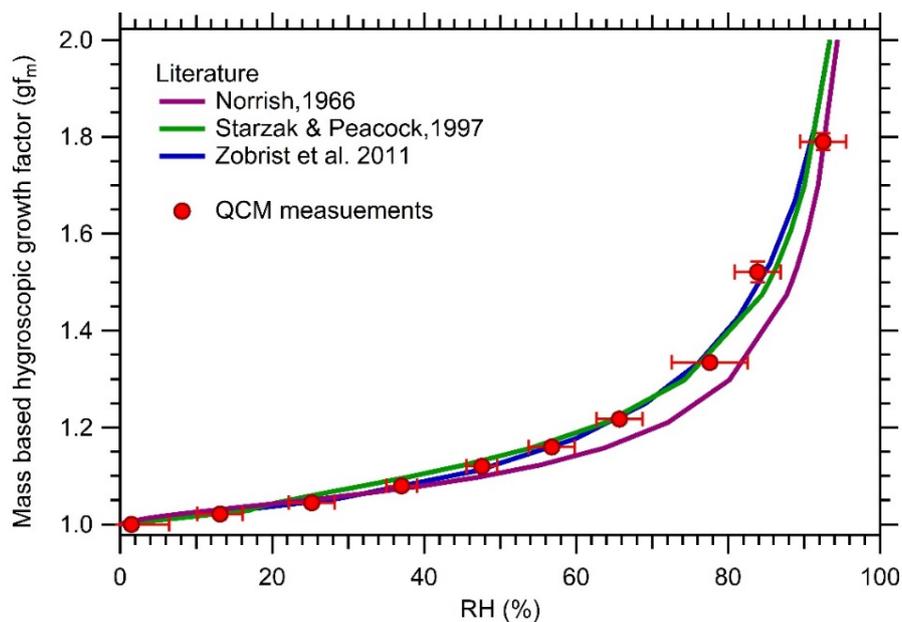


Figure S3. Mass-based hygroscopic growth factor gf_m of amorphous sucrose particles at different RH values in the subsaturated region obtained from the quartz crystal microbalance (QCM) measurements. The experimental values strictly follow the gf_m values retrieved using different models (Norrish, Starzak and Peacock, and Zobrist) from the literature.

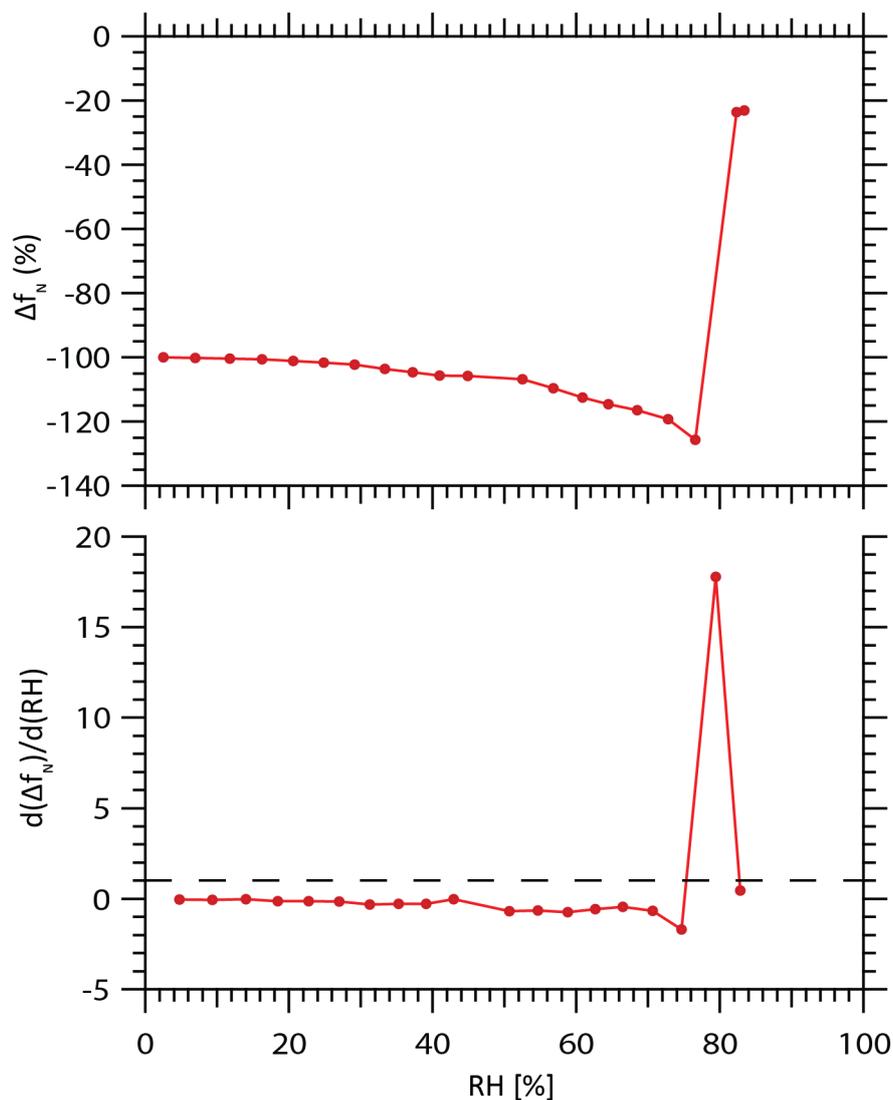


Figure S4. The deliquescence phase transition behavior of laboratory-generated $(\text{NH}_4)_2\text{SO}_4$ particles. Δf_N represents the change in the oscillation frequency of the quartz crystal microbalance (QCM) sensor resulting due to water uptake by the particles at different RH conditions normalized to that of the dry particles at $\text{RH} < 5\%$, expressed as percentage. The decrease in the value of Δf_N indicates the water uptake at different RH conditions in the sub-saturated region. The derivative of Δf_N with respect to RH, $d(\Delta f_N)/d(\text{RH})$ is plotted against different RH conditions and the respective RH values for $d(\Delta f_N)/d(\text{RH}) \geq 1$, marked by the black dotted line, represent the deliquescence relative humidity (DRH) range (76-80%) for the $(\text{NH}_4)_2\text{SO}_4$ particles.

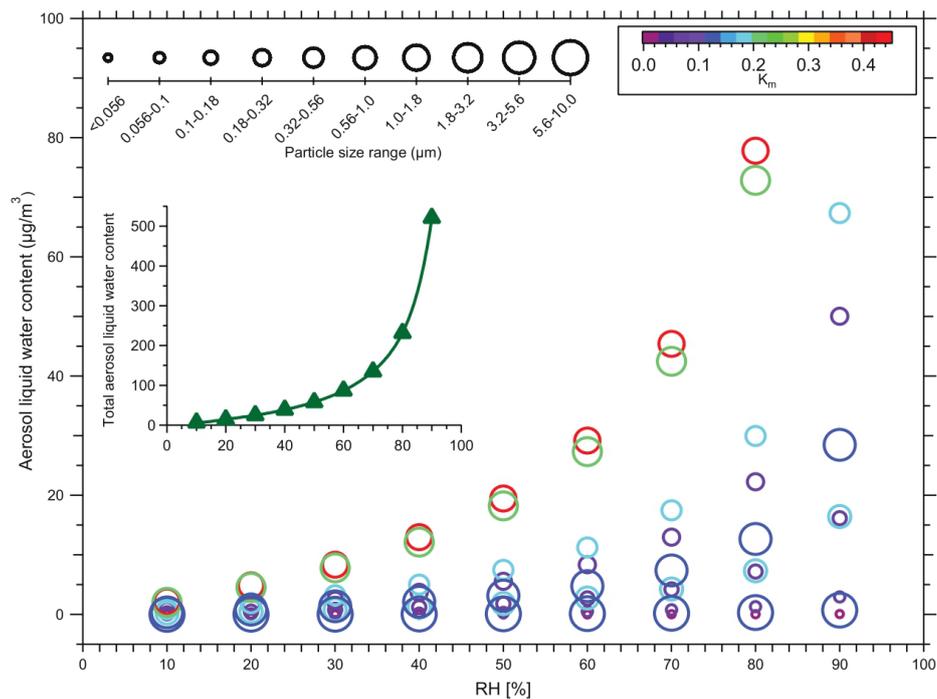


Figure S5. Aerosol Liquid Water Content (ALWC) in size-resolved ambient aerosol particles from Munnar across varied relative humidity (RH) conditions in the subsaturated regime. ALWC under different RH conditions is estimated by integrating size-resolved hygroscopic growth factor values with particle number size distribution (PNSD) measurements, assuming volume conservation during aerosol water uptake processes. The marker sizes correspond linearly to particle size, while marker colors signify the mean value of the mass-based hygroscopicity parameter (κ_m) for each size-range, indicated by the color palette. The total ALWC averaged over all particles below 10 μm in size is also shown in inset.

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