

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

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KEYPOINTS

- QCM technique revealed aerosol properties at low RH values offering insights that may not be captured by traditional size-based measurements
- The size-dependent variations in aerosol properties below DRH emphasize the need to understand how aerosol properties change with RH history
- Organic in aerosols complicate its cloud forming ability, necessitating additional studies in diverse environments to improve climate models

ABSTRACT

Comprehending the intricate interplay between atmospheric aerosols and water vapour in subsaturated regions is vital for accurate modelling of aerosol–cloud–radiation–climate dynamics. But the microphysical mechanisms governing these interactions with ambient aerosols remain inadequately understood. Here we report results from high-altitude, relatively pristine site in Western-Ghats of India during monsoon, serving as a baseline for climate processes in one of the world’s most polluted regions. Utilizing a novel quartz crystal microbalance (QCM) approach, we conducted size-resolved sampling to analyse humidity-dependent growth factors, hygroscopicity, deliquescence behaviour, and aerosol liquid water content (ALWC). Fine-mode aerosols ($\leq 2.5 \mu\text{m}$) exhibited size-dependent interactions with water vapour, contributing significantly to ALWC. Deliquescence was observed in larger aerosols ($> 180 \text{ nm}$), influenced by organic species, with deliquescence relative humidity (DRH) lower than that of pure inorganic salts. This research highlights the significance of understanding ambient aerosol-water interactions and hygroscopicity for refining climate models in subsaturated conditions.

PLAIN LANGUAGE SUMMARY

Aerosol particles interact with water vapour in the atmosphere. Understanding these interactions in sub – and super-saturated regions is crucial because they affect processes such as cloud formation, radiation, and climate. We collected ambient aerosol samples from Western Ghats in India to understand natural processes that happen without significant human interventions. Using a special technique involving a quartz crystal sensor, we measured different aspects, such as how aerosols grow with humidity, their ability to uptake the water molecules, and when they transition from solid to liquid (deliquescence). Interestingly, fine particles, especially those smaller than $2.5 \mu\text{m}$, exhibited unique behaviours. While larger particles underwent a transition from solid to liquid under certain conditions, this didn’t happen for the smaller particles. These findings highlight the importance of understanding these interactions for more accurate climate predictions.

1. INTRODUCTION

Understanding the hygroscopicity of atmospheric aerosols is crucial for assessing cloud formation and their climate and air pollution impact (Cheung et al., 2015). Investigation of the hygroscopicity of ambient aerosols poses significant challenges due to their chemical complexity, particle size variation, phase state, and viscosity. Techniques like Fourier transform infrared spectroscopy (FTIR) (Y. Liu et al., 2008; Y. Liu & Laskin, 2009), quartz crystal microbalance (QCM) (Chao et al., 2020; Demou et al., 2003; P. Liu et al., 2016a; P. Liu, Song, et al., 2018a), Raman spectroscopy (Ling & Chan, 2008; Y. J. Liu et al., 2008), electrodynamic balance (EDB) (Choi & Chan, 2002; Peng & Chan, 2001; Pope et al., 2010), optical microscopy (OM) (Ahn et al., 2010; Eom et al., 2014; Gupta et al., 2015), hygroscopicity tandem differential mobility analysis (HTDMA) (Cheung et al., 2015; Prenni et al., 2007; Zieger et al., 2017a) and size-selected cloud condensation nuclei (CCN) spectrometry (Petters et al., 2007; Pöhlker et al., 2016; Rose et al., 2008) have been employed to study the hygroscopicity of laboratory – generated and ambient aerosols (Tang et al., 2019). However, a consensus on the most effective method is lacking, leading to inconsistent and incomparable results. Hygroscopicity measurements primarily focus on accumulation mode particles, limiting data on nucleation and coarse mode particles with complex behaviours. Discrepancies in hygroscopic properties and organic and inorganic species composition in atmospheric aerosols result in diverse growth factors and phase transitions, crucial for accurate climate modelling (Li et al., 2021).

The QCM is effective in determining mass-based hygroscopicity and physical property variations of atmospherically-relevant aerosols (Demou et al., 2003; P. Liu et al., 2016b; P. Liu, Song, et al., 2018b). It overcomes the limitations of traditional methods like HTDMA, which has a narrow particle size range with limited relative humidity (RH) resolution (Zhao et al., 2022). Conventional HTDMA methods inadequately reveal hygroscopicity heterogeneity and phase transitions in wider aerosol size range, impacting processes like CCN activation and aerosol liquid water (Li et al., 2021). QCM's mass-based measurements accurately quantify aerosol hygroscopicity behaviour across a wide RH range in the atmosphere, detailing physical property variations during phase transitions and help to delineate water uptake mechanisms, including solubility and water diffusion limitations (P. Liu, Li, et al., 2018; P. Liu, Song, et al., 2018b).

India's climate is distinct and intricate, and as per the 2020 climate change assessment report, India has faced challenges like temperature rise and extreme weather events since the mid-20th century. A key hurdle in addressing these issues is the absence of systematic aerosol characteristic measurements, particularly in aerosol-water vapor interactions in the subsaturated regime (Cheung et al., 2015). Aerosol particles from a forest-surrounded by pristine area are anticipated to contain substantial biogenic organic aerosols. The dynamic chemical transformation of secondary organic aerosols (SOA) in the atmosphere leads to intricate compositions. Atmospheric processing further complicates the understanding of phase state and properties of atmospheric aerosols (Rastak et al., 2017). In this study, QCM measurements are utilized to explore aerosol hygroscopicity dependence on RH, chemical composition, and size across a broad range of ambient aerosols in India's relatively pristine setting.

2. MATERIALS AND METHODS

Size-resolved ambient aerosols were sampled at the Natural Aerosol and Bioaerosol High Altitude (NABHA) Laboratory at the College of Engineering, Munnar (10.0930° N, 77.0682° E; 1600 m above mean sea level), a high-altitude and typically clean site situated in the Western Ghats of India, using a 10-stage micro-orifice uniform deposit impactor (MOUDI-II 120-R, TSI) (V. Marple et al., 2014; V. A. Marple et al., 1991) during the monsoon season (August-September) of 2021. Ambient particles collected on PTFE (Polytetrafluoroethylene) filters were transferred to a hydrophobic SiO₂-coated quartz sensor by gently pressing the filter paper onto the sensor. The hygroscopic growth factor, hygroscopicity parameter and deliquescence relative humidity (DRH) were estimated using a highly sensitive mass balance instrument, QCM (QSense Analyzer, Biolin Scientific) (P. Liu, Song, et al., 2018a; Reviakine et al., 2011). The accuracy and robustness of the method were demonstrated by performing measurements of the hygroscopic growth factor and DRH for sucrose and (NH₄)₂SO₄ particles, respectively, which were compared with previously reported results (Arenas et al., 2012a; Chao et al., 2020; Martin, 2000; Norrish, 1966; Peng et al., 2022; Starzak & Peacock, 1997; Zobrist et al., 2011) (Fig. S3 and S4). The hygroscopic growth factor and the corresponding hygroscopicity parameter, κ , were determined over a wide range of RH conditions using κ -Köhler theory (Petters & Kreidenweis, 2007), where κ represents a quantitative measure of aerosol water uptake characteristics and CCN activity (Zhao et al., 2022). The water uptake characteristics and the phase transition behaviours of the ambient

samples were investigated and compared between different size ranges of particles below 10 μm collected using the MOUDI sampler. The Aerosol Liquid Water Content (ALWC) (Bian et al., 2014) at different RH conditions was also estimated from corresponding hygroscopicity parameters for each size range of particles (Fig. S5). The number size distributions of the ambient aerosol particles of size 10 to 420 nm [measured in parallel using a scanning mobility particle sizer – SMPS, comprised of an electrostatic classifier (EC, TSI Model 3082) equipped with a differential mobility analyser (DMA, TSI Model 3081) and a condensation particle counter (CPC, TSI Model 3750)], were then converted to mass size distributions assuming a particle density of 1.2 g cm^{-3} (DeCarlo et al., 2004). The supermicron particle mass size distribution for the same season, but a different year, was used in this study, which had been obtained using an Ultraviolet Aerodynamic Particle Sizer (UV-APS, TSI Inc., model 3314) (Valsan et al., 2016).

Meteorological parameters were recorded using an automatic weather station (AWS, Climate Sensor US) during the sampling period. Figure 1 shows the sampling site – Munnar, along with aerosol optical depth (AOD) over India during the monsoon season, indicating the relatively pristine nature of the sampling site. During the measurement, airmasses predominantly arrived from the southwest direction, bringing clean marine influx to the observational site and resulting in a low influence of anthropogenic emissions (Fig. 1). A more detailed description of the instruments, experimental techniques, and estimation of various parameters is provided in the Supporting Information (SI).

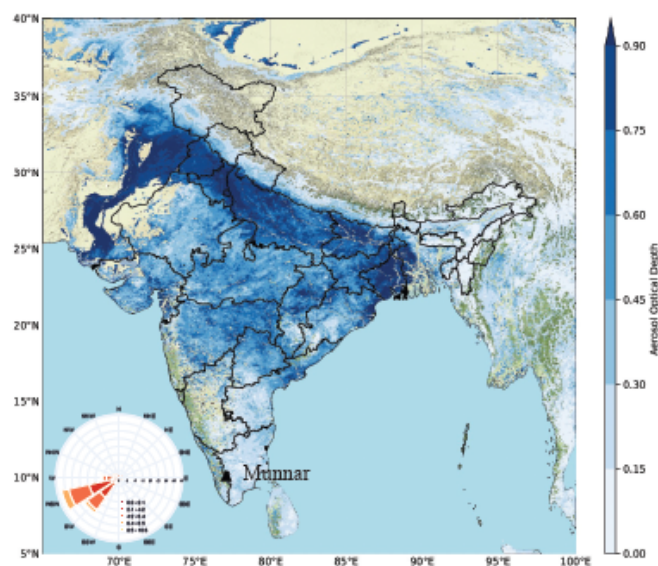


Figure 1. Spatial distribution of average Aerosol Optical Depth (AOD) derived from MODIS (Moderate Resolution Imaging Spectroradiometer) Level 2 data over the Indian continental region during the monsoon season of 2021 (June - September). The AOD distribution clearly indicates a relatively low aerosol loading over the observational site of Munnar (marked as a black triangle) compared with the other parts of India. The wind rose diagram shown in the inset shows the average wind speed and wind direction arriving at the sampling site during the ambient aerosol sampling period (August – September 2021). The prevailing air masses mostly originated over the Indian Ocean and arrived from southwest direction, bringing clean marine influx to the observational site, confirming the relatively low influence of anthropogenic activities.

3. RESULTS AND DISCUSSION

The QCM sensor's oscillation frequency variation (Δf) signifies water uptake and release, providing insights into adsorption, desorption, and physical states of particles during solid-to-aqueous phase transition (Arenas et al., 2012b). Normalizing the frequency shift at higher RH to that of deposited dry sample (RH <5%) yields the percentage value, Δf_N (Chao et al., 2020). Figure 2 shows Δf_N for the ambient aerosol particles across various RH levels and sampled size ranges. A negative sign in Δf_N indicates frequency reduction with increasing RH due to water uptake. Further details of Δf_N and its derivative, $d(\Delta f_N)/(RH)$, are available in SI.

For the particle size <180 nm (Fig. 2a), Δf_N decreased, implying water uptake, with <56 nm particles exhibiting the lowest value. In Figure 2b and 2c, particles showed increased water uptake at lower RH, evidenced by Δf_N decrease. However, Δf_N suddenly increased at specific RH, indicating a drastic water uptake leading to deliquescence-induced phase transition. Figures 2a-c revealed deliquescence in particles >180 nm at different RH (known as DRH), absent in those <180 nm. Past studies reported no deliquescence for ambient and laboratory-generated atmospherically relevant particles even at high RH, attributing them to organic nature (Arenas et al., 2012a; Brooks et al., 2002; Chao et al., 2020; Peng et al., 2022). We hypothesize that particles <180 nm were likely dominated by freshly formed secondary organic aerosols (SOA) from biogenic volatile organic compound (VOC) oxidation, supported by the densely vegetated/forest region and season. Concurrent quasi-continuous measurements during the same campaign indicated upto ~90% organic fraction in NR-PM₁ (non-refractory particulate matter with an aerodynamic diameter $\leq 1 \mu\text{m}$; Tab. S2) with details discussed in subsequent studies.

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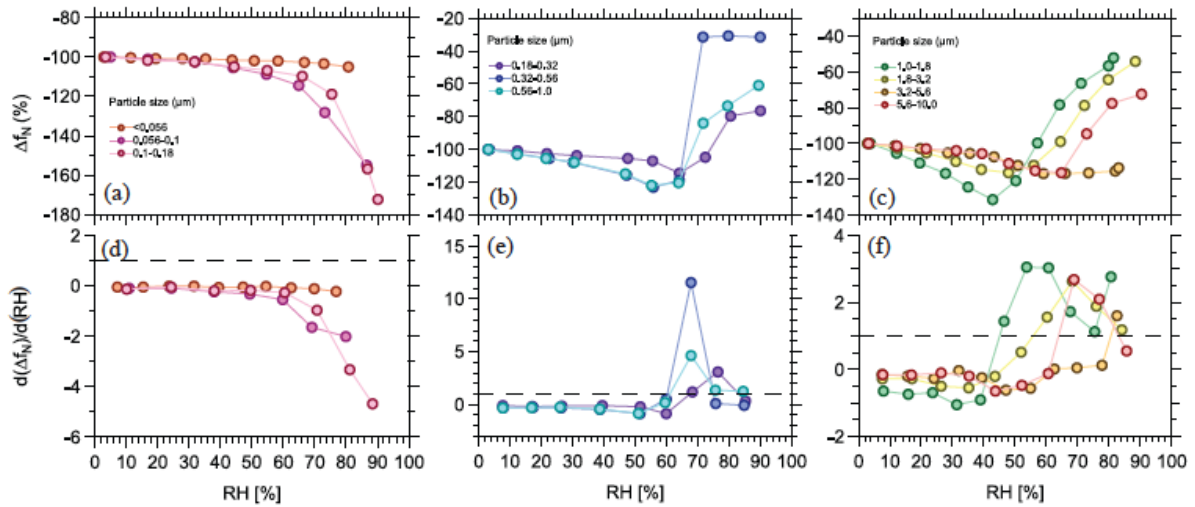


Figure 2. Deliquescence phase transition behaviour of size-resolved ambient aerosol particles from Munnar. For panels a, b, and c, Δf_N represents the change in the oscillation frequency of the quartz crystal microbalance (QCM) sensor resulting due to water uptake by the ambient aerosol particles at different relative humidity (RH) conditions normalized to that of the dry aerosol particles at RH <5%, expressed as percentage. The decrease in the value of Δf_N for each size range for the sampled ambient aerosol particles indicates the water uptake at different RH conditions in the subsaturated regime. The solid markers and lines identify different particle size ranges. In panels d, e, and f, the derivative of Δf_N with respect to RH ($d(\Delta f_N)/d(RH)$) is plotted against RH to determine the deliquescence relative humidity (DRH) value corresponding to the respective aerosol size ranges. The RH values at which $d(\Delta f_N)/d(RH)$ becomes ≥ 1 (marked by the dotted line) represent the DRH values for the individual aerosol size ranges.

Following Chao et al. (2020), DRH, indicated by $d(\Delta f_N)/d(RH) \geq 1$ ranged from ~60 – 68% for three size ranges (180 nm – 320 nm; 320 nm – 560 nm; 560 nm – 1 μ m) in this study (Fig. 2e). Above 180 nm, inorganic salts, like $(NH_4)_2SO_4$, may contribute to DRH appearance (Hu et al., 2010) (Fig. 2b,e). These DRH values, smaller than pure $(NH_4)_2SO_4$ (DRH=80%), suggest mixed salts and/or organic compounds in the sampled aerosol particles. The deliquescence behaviour of organic-inorganic mixtures introduces complexity due to solubility limitations, influenced by the organic species in the aerosol particles. Previous studies noted reduced DRH of inorganic species and sometimes, unaffected by organics (Smith et al., 2011, 2012, 2013). Thus, we hypothesize 180 nm to 1 μ m particles at this site reflect a complex mixture of inorganic salts like $(NH_4)_2SO_4$ and water-soluble organic compounds. Supermicron particles (>1 μ m), except 1 – 1.8 μ m, showed <17% Δf_N , indicating lower water uptake (Fig. 2c). Beyond maximum water uptake, the three size ranges exhibited different DRH values. 1.8 – 3.2 μ m and 5.6 – 10 μ m exhibited DRH between 56 – 68%, suggesting a mixture of water-soluble organic compounds and inorganic salts. However, 3.2 – 5.6 μ m showed a very high DRH (>81%) with the organic fraction minimally affecting DRH due to solubility limitations. For 1.8 – 3.2 μ m and 5.6 – 10 μ m, reduced DRH

compared to pure inorganic salts suggests a mix of water-soluble organic and inorganic compounds (Smith et al., 2011, 2012, 2013). The 1.0 – 1.8 μm range exhibited a $>30\%$ Δf_N reduction at $\sim 42\%$ RH, indicating a higher water uptake and the observed DRH appeared to be $\sim 45 - 53\%$, consistent with the highest κ_m (~ 0.44 , see discussion below) among all measured size ranges (Chao et al., 2020). The low DRH at this site suggests presence of highly water-soluble organic material within the organic-inorganic (sea salt) mixture in this size range.

Figure 3 shows the size-resolved mass-based hygroscopic growth factors (gf_m) and the corresponding mass-based hygroscopicity parameters (κ_m) of ambient aerosol particles calculated from QCM measurements (Text S4 and S5) over a wide RH range (2 – 90%). In Figure 3a, the growth factor increases with water uptake in humid air, showing size-dependent variations across different RH levels (indicated by markers). Each size range had a maximum RH limit beyond which gf_m estimation was impossible due to deliquescence, as observed in previous studies (Chao et al., 2020). Accordingly, the highest $gf_m=1.72$ was recorded at RH=90% for the 100 – 180 nm particles. The pronounced variations in gf_m between different-sized particles at a given RH indicate varying chemical composition and hygroscopicity across the sampled sizes.

Figure 3b shows the size-dependent average κ_m values, calculated from the gf_m values in Figure 3a. Each particle size range exhibited minimal κ_m variation (average $\kappa_m \pm 0.007$), suggesting ideal solution behaviour across the wide RH range (Z. Wang et al., 2017). The average κ_m values (indicated by dashed lines in Figure 3b) of the size-resolved ambient particles varied significantly between 0.016 (<0.056 nm) and 0.44 (1.0 – 1.8 μm) over the investigated RH range. This implies κ_m as a function of size, and indicates distinct chemical compositions for each size fraction. For example, the small increase in κ_m from 0.016 to 0.078 for particles <56 nm to 56-100 nm, respectively, is potentially owing to aging processes (Zhang et al., 2023). Aerosol particles <320 nm exhibited very low average κ_m (0.06), which may be primarily due to the strong dominance of organic compounds in these size ranges (Demou et al., 2003; Li et al., 2021; Shi et al., 2022). The particles in the size range of 320 nm – 1 μm were moderately hygroscopic (average $\kappa_m=0.19$) indicating the influence of inorganic salts (Wu et al., 2016). The particles between 1.0 – 1.8 μm exhibited highest κ_m (0.44), which implies the presence of highly hygroscopic material such as sea salt

(Zieger et al., 2017b) mixed with organic compounds. Even larger particles exhibited reduced κ_m , possibly due to the presence of dust particles (Koehler et al., 2009), transported at this site during monsoon season (Valsan et al., 2016). The κ -Köhler parameterization fit accurately elucidates the gf_m within each size range over a wide range of RH values (Petters & Kreidenweis, 2007).

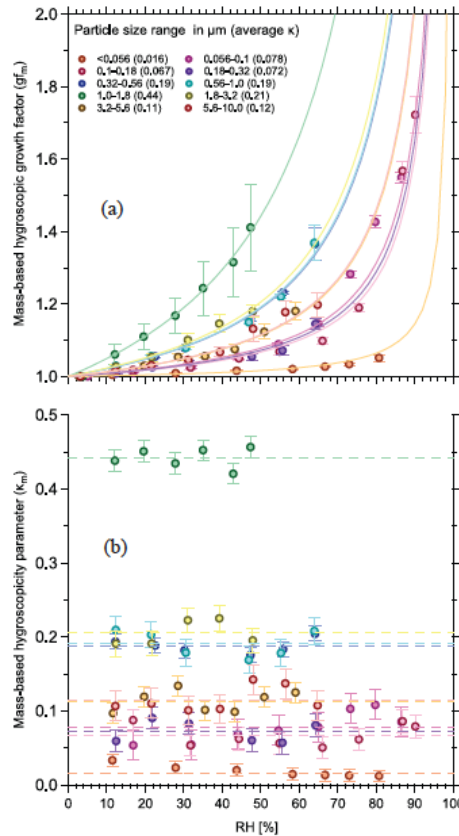


Figure 3. Size-resolved hygroscopicity measurements of ambient aerosol particles at the high-altitude site, Munnar, during the Monsoon season (August-September 2021). (a) Mass-based hygroscopic growth factor (gf_m) derived using a quartz crystal microbalance (QCM) for ten different size ranges of ambient aerosol particles at different relative humidity (RH) conditions in the subsaturated regime (circles). The solid lines represent the corresponding κ -Köhler growth factor fits obtained using the mean value of mass-based hygroscopicity parameters, κ_m . The error bars represent the variations in gf_m averaged over the mass change corresponding to different overtone frequencies of the QCM sensor at respective RH conditions. The values in parentheses are the mean κ_m values corresponding to the respective size ranges. (b) The data points are the κ_m values calculated based on the gf_m (as shown in (a)) using the κ -Köhler theory for different RH conditions in the subsaturated regime. The dotted lines represent the mean κ_m value for each size range of ambient aerosol particles and the error bars represent one standard deviation.

Figure 4 shows the size dependency of κ_m in the subsaturated regime measured by QCM together with κ_{CCN} (Text S2) in the supersaturated regime determined by size-resolved CCN measurements and aerosol size distributions obtained by SMPS and UV-APS. The submicron particles show a bimodal number size distribution with an Aitken mode peak at 63 nm and an accumulation mode at 145 nm, while the mass size distributions peak at 350 nm and 3 μm ,

respectively. The average κ_{CCN} increased from a value of 0.28 for the Aitken mode particles to 0.47 for the accumulation mode particles. In the submicron region, κ_{m} exhibited a nominal increase with particle size (Wu et al., 2016). in parallel with the increase in κ_{CCN} , albeit at much lower absolute values. This increase likely reflects the decrease of organic fraction with size in the submicron range, which is supported by the appearance of deliquescence in the size range above 180 nm, indicating the presence of inorganic salts. In the supermicron region (for particles $\geq 1 \mu\text{m}$), κ_{m} peaked due to the potential presence of NaCl particles in the size range 1.0 – 1.8 μm , and decreased again in the higher size ranges ($>1.8 \mu\text{m}$) likely because of the presence of dust particles. The average κ_{m} value (0.18) obtained from QCM measurements across all the sampled size ranges in the subsaturated regime was lower than the average κ_{CCN} (0.39), likely due to the solubility limitation (Dusek et al., 2011; Hersey et al., 2013; Rastak et al., 2017; Riipinen et al., 2015; Wittbom et al., 2018) of organic and inorganic compounds in ambient aerosol particles below the DRH. Most inorganic species are completely dissolved beyond the DRH point in the subsaturated regime, allowing κ_{CCN} to assume complete particle solubility (Pajunoja et al., 2015; Petters & Kreidenweis, 2007).

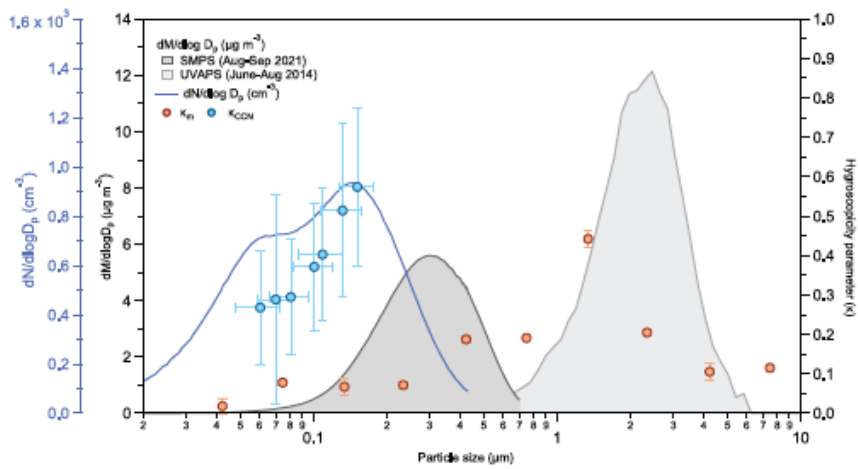


Figure 4. Aerosol size distributions, hygroscopicity parameters derived from quartz crystal microbalance (QCM) experiments (κ_{m}), and from size-resolved cloud condensation nuclei (CCN) measurements (κ_{CCN}). The number size distribution obtained using a Scanning Mobility Particle Sizer (SMPS) over the size range of 10 – 430 nm (blue curve) was measured during the sampling period and exhibited a bimodal distribution. The mass size distributions (dark grey shaded area) were derived based on the aerosol number size distribution by assuming a density of 1.2 g cm^{-3} for the submicron region. The mass size distribution for the supermicron range (light grey shaded area) was obtained using Ultraviolet Aerodynamic Particle Sizer (UV-APS) measurements during the same season (June–August) but for a different year (2014). The hygroscopicity parameters derived from QCM experiments (κ_{m} ; orange points) and size-resolved CCN measurements (κ_{CCN} ; blue points) are shown for the comparison. The error bars for κ_{m} and κ_{CCN} indicate the measurement uncertainty and variability, respectively.

Unlike QCM measurements, the hygroscopicity parameters obtained in previous studies using the HTDMA technique (κ_{HTDMA}) are often consistent with κ_{CCN} , as the particle

hygroscopicity is determined by particle size increase above DRH point, unaffected by solubility limits (Pajunoja et al., 2015; Y. Wang et al., 2018; Wu et al., 2013). The technical limitations, particularly limited diameter changes corresponding to gf_m values less than 2.0, prevent HTDMA from measuring aerosol hygroscopic properties below the DRH point in the subsaturated regime (Laskina et al., 2015). In contrast, the QCM technique, which is not subject to these limitations, can be used to understand the water uptake characteristics of aerosol particles in the subsaturated regime below DRH and substantially enhance our understanding about solubility limitations of organic and inorganic compounds in the lower RH region. Such an enhancement in the knowledge is crucial for accurately quantifying the radiative forcing effects of ambient aerosol particles in the atmosphere. The strong size dependence of κ_m also indicated varying chemical composition for different size ranges measured in this study.

4. SUMMARY AND ATMOSPHERIC IMPLICATIONS

We report the first results based on a high-sensitivity QCM technique to investigate the mass-based growth factor and hygroscopicity parameter of size-resolved ambient aerosols over a wide range of RH from a relatively pristine high-altitude site in India. For the investigated size ranges, the pronounced variations in gf_m below the DRH provided an opportunity to better understand the changes in ambient aerosol properties even at low RH values, which may not be revealed by size-based growth factor measurements (Hu et al., 2010; Laskina et al., 2015). These measurements clearly highlight the important and critical role in knowing the changes in aerosol properties based on their RH history for an improved understanding of water uptake, phase transition, and radiative impact of atmospheric aerosol particles (Zhao et al., 2022). Based on the estimated κ_m values, the ambient aerosol particles exhibited the behaviour of an ideal solution (Pajunoja et al., 2015) and strong size-dependent chemical composition. We further observed a pronounced size dependency of the DRH values potentially resulting from complexities of organic solubility in organic-inorganic mixtures (Li et al., 2021). Such a complexity owing to the presence of various organic species may alter the CCN behaviour of atmospheric aerosol particles due to phase transitions and changes in physical properties. The understanding of CCN activation in the supersaturated regime under the assumption of complete solubility of particles is relatively well established. But the understanding of the thermodynamic properties associated with particle growth in the subsaturated regime still remains a challenge. Our findings emphasize the need for additional experiments on ambient and atmospherically relevant laboratory-generated aerosol particles

387 using high-sensitivity techniques like QCM. We demonstrated the importance of this
388 technique to better understand the rapid changes in aerosol properties resulting from exposure
389 to a wide range of atmospheric RH conditions. The enhancement in our understanding of the
390 complex interplay between water vapour and aerosol particles will help in developing more
391 accurate models to effectively describe the role of aerosols in atmospheric processes to
392 reduce climate uncertainties and assess the impact of air pollution on human and ecosystem
393 health.

ASSOCIATED CONTENT

Author contributions

SSG conceived the idea. SSG and PL conceptualized the study and designed the research. CJ further developed and validated the QCM measurements for the ambient aerosol studies. CJ performed the field measurement campaign to collect the aerosol samples with support from AS, KNK, and RKA. CJ performed all the laboratory experiments using QCM with support from SS. GVJ performed the satellite data analysis to obtain the AOD values. CJ carried out the data analysis obtained from QCM with input from PL and SSG. CJ and AS performed the scientific interpretation of the QCM data under the mentorship of RR, SSG and PL. CJ wrote the first draft of manuscript under the mentorship of SSG with inputs from PL and RR, and further edits from SY. MOA and STM further provided critical and valuable inputs on the manuscript.

Open research

The data used in the manuscript has been deposited in an open research repository as Excel files, accessible at <https://doi.org/10.6084/m9.figshare.24512377>. MODIS data were sourced from the Level 2 and Atmosphere Archive and Distribution System (LAADS) (https://doi.org/10.5067/MODIS/MYD04_3K.061). Figures were prepared using Igor Pro version 9 (WaveMetrics Inc.), licensed by SSG and accessible at <https://www.wavemetrics.com/software/igor-pro-9>.

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