

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

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26 **KEYPOINTS**

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- QCM technique revealed aerosol properties at low RH values offering insights that
28 may not be captured by traditional size-based measurements
 - The size-dependent variations in aerosol properties below DRH emphasize the need to
29 understand how aerosol properties change with RH history
 - Organic in aerosols complicate its cloud forming ability, necessitating additional
30 studies in diverse environments to improve climate models
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32

33 **ABSTRACT**

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

48 **PLAIN LANGUAGE SUMMARY**

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

60 1. INTRODUCTION

61 Understanding the hygroscopicity of atmospheric aerosols is crucial for assessing cloud
62 formation and their climate and air pollution impact (Cheung et al., 2015). Investigation of
63 the hygroscopicity of ambient aerosols poses significant challenges due to their chemical
64 complexity, particle size variation, phase state, and viscosity. Techniques like Fourier
65 transform infrared spectroscopy (FTIR) (Y. Liu et al., 2008; Y. Liu & Laskin, 2009), quartz
66 crystal microbalance (QCM) (Chao et al., 2020; Demou et al., 2003; P. Liu et al., 2016a; P.
67 Liu, Song, et al., 2018a), Raman spectroscopy (Ling & Chan, 2008; Y. J. Liu et al., 2008),
68 electrodynamic balance (EDB) (Choi & Chan, 2002; Peng & Chan, 2001; Pope et al., 2010),
69 optical microscopy (OM) (Ahn et al., 2010; Eom et al., 2014; Gupta et al., 2015),
70 hygroscopicity tandem differential mobility analysis (HTDMA) (Cheung et al., 2015; Prenni
71 et al., 2007; Zieger et al., 2017a) and size-selected cloud condensation nuclei (CCN)
72 spectrometry (Petters et al., 2007; Pöhlker et al., 2016; Rose et al., 2008) have been employed
73 to study the hygroscopicity of laboratory – generated and ambient aerosols (Tang et al.,
74 2019). However, a consensus on the most effective method is lacking, leading to inconsistent
75 and incomparable results. Hygroscopicity measurements primarily focus on accumulation
76 mode particles, limiting data on nucleation and coarse mode particles with complex
77 behaviours. Discrepancies in hygroscopic properties and organic and inorganic species
78 composition in atmospheric aerosols result in diverse growth factors and phase transitions,
79 crucial for accurate climate modelling (Li et al., 2021).

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

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93 India's climate is distinct and intricate, and as per the 2020 climate change assessment report,
94 India has faced challenges like temperature rise and extreme weather events since the mid-
95 20th century. A key hurdle in addressing these issues is the absence of systematic aerosol
96 characteristic measurements, particularly in aerosol-water vapor interactions in the
97 subsaturated regime (Cheung et al., 2015). Aerosol particles from a forest-surrounded by
98 pristine area are anticipated to contain substantial biogenic organic aerosols. The dynamic
99 chemical transformation of secondary organic aerosols (SOA) in the atmosphere leads to
100 intricate compositions. Atmospheric processing further complicates the understanding of
101 phase state and properties of atmospheric aerosols (Rastak et al., 2017). In this study, QCM
102 measurements are utilized to explore aerosol hygroscopicity dependence on RH, chemical
103 composition, and size across a broad range of ambient aerosols in India's relatively pristine
104 setting.

105 **2. MATERIALS AND METHODS**

106 Size-resolved ambient aerosols were sampled at the Natural Aerosol and Bioaerosol High
107 Altitude (NABHA) Laboratory at the College of Engineering, Munnar (10.0930° N, 77.0682°
108 E; 1600 m above mean sea level), a high-altitude and typically clean site situated in the
109 Western Ghats of India, using a 10-stage micro-orifice uniform deposit impactor (MOUDI-II
110 120-R, TSI) (V. Marple et al., 2014; V. A. Marple et al., 1991) during the monsoon season
111 (August-September) of 2021. Ambient particles collected on PTFE (Polytetrafluoroethylene)
112 filters were transferred to a hydrophobic SiO₂-coated quartz sensor by gently pressing the
113 filter paper onto the sensor. The hygroscopic growth factor, hygroscopicity parameter and
114 deliquescence relative humidity (DRH) were estimated using a highly sensitive mass balance
115 instrument, QCM (QSense Analyzer, Biolin Scientific) (P. Liu, Song, et al., 2018a; Reviakine
116 et al., 2011). The accuracy and robustness of the method were demonstrated by performing
117 measurements of the hygroscopic growth factor and DRH for sucrose and (NH₄)₂SO₄
118 particles, respectively, which were compared with previously reported results (Arenas et al.,
119 2012a; Chao et al., 2020; Martin, 2000; Norrish, 1966; Peng et al., 2022; Starzak & Peacock,
120 1997; Zobrist et al., 2011) (Fig. S3 and S4). The hygroscopic growth factor and the
121 corresponding hygroscopicity parameter, κ , were determined over a wide range of RH
122 conditions using κ -Köhler theory (Petters & Kreidenweis, 2007), where κ represents a
123 quantitative measure of aerosol water uptake characteristics and CCN activity (Zhao et al.,
124 2022). The water uptake characteristics and the phase transition behaviours of the ambient

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

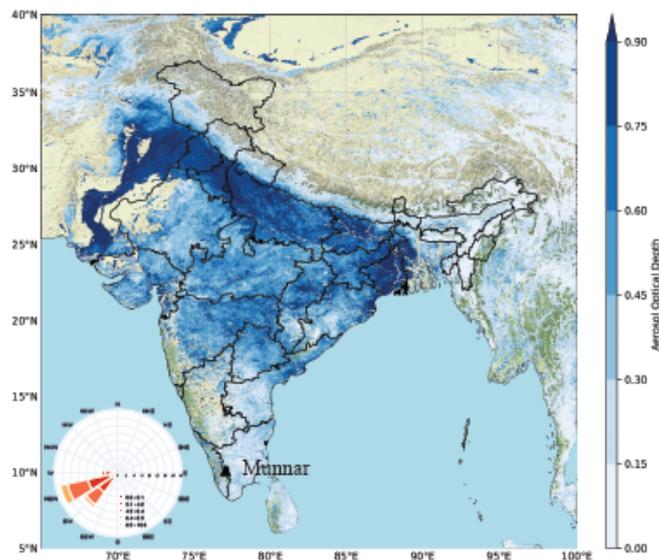
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138 Meteorological parameters were recorded using an automatic weather station (AWS, Clima
 139 Sensor US) during the sampling period. Figure 1 shows the sampling site – Munnar, along
 140 with aerosol optical depth (AOD) over India during the monsoon season, indicating the
 141 relatively pristine nature of the sampling site. During the measurement, airmasses
 142 predominantly arrived from the southwest direction, bringing clean marine influx to the
 143 observational site and resulting in a low influence of anthropogenic emissions (Fig. 1).

144 A more detailed description of the instruments, experimental techniques, and estimation of
 145 various parameters is provided in the Supporting Information (SI).

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

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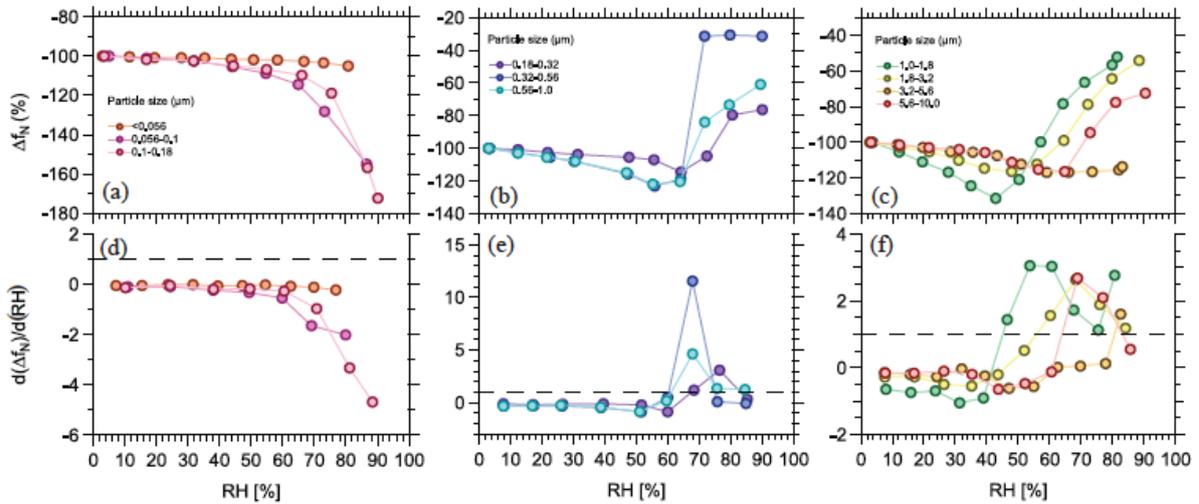
175 **3. RESULTS AND DISCUSSION**

176 The QCM sensor's oscillation frequency variation (Δf) signifies water uptake and release,
177 providing insights into adsorption, desorption, and physical states of particles during solid-to-
178 aqueous phase transition (Arenas et al., 2012b). Normalizing the frequency shift at higher RH
179 to that of deposited dry sample (RH <5%) yields the percentage value, Δf_N (Chao et al.,
180 2020). Figure 2 shows Δf_N for the ambient aerosol particles across various RH levels and
181 sampled size ranges. A negative sign in Δf_N indicates frequency reduction with increasing RH
182 due to water uptake. Further details of Δf_N and its derivative, $d(\Delta f_N)/(RH)$, are available in SI.

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

198



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

209

210 Following Chao et al. (2020), DRH, indicated by $d(\Delta f_N)/d(\text{RH}) \geq 1$ ranged from $\sim 60 - 68\%$ for
 211 three size ranges (180 nm – 320 nm; 320 nm – 560 nm; 560 nm – 1 μm) in this study (Fig.
 212 2e). Above 180 nm, inorganic salts, like $(\text{NH}_4)_2\text{SO}_4$, may contribute to DRH appearance (Hu
 213 et al., 2010) (Fig. 2b,e). These DRH values, smaller than pure $(\text{NH}_4)_2\text{SO}_4$ (DRH=80%),
 214 suggest mixed salts and/or organic compounds in the sampled aerosol particles. The
 215 deliquescence behaviour of organic-inorganic mixtures introduces complexity due to
 216 solubility limitations, influenced by the organic species in the aerosol particles. Previous
 217 studies noted reduced DRH of inorganic species and sometimes, unaffected by organics
 218 (Smith et al., 2011, 2012, 2013). Thus, we hypothesize 180 nm to 1 μm particles at this site
 219 reflect a complex mixture of inorganic salts like $(\text{NH}_4)_2\text{SO}_4$ and water-soluble organic
 220 compounds. Supermicron particles ($>1\ \mu\text{m}$), except 1 – 1.8 μm , showed $<17\%$ Δf_N , indicating
 221 lower water uptake (Fig. 2c). Beyond maximum water uptake, the three size ranges exhibited
 222 different DRH values. 1.8 – 3.2 μm and 5.6 – 10 μm exhibited DRH between 56 – 68%,
 223 suggesting a mixture of water-soluble organic compounds and inorganic salts. However, 3.2
 224 – 5.6 μm showed a very high DRH ($>81\%$) with the organic fraction minimally affecting
 225 DRH due to solubility limitations. For 1.8 – 3.2 μm and 5.6 – 10 μm , reduced DRH

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

233

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

244

245 Figure 3b shows the size-dependent average κ_m values, calculated from the gf_m values in
246 Figure 3a. Each particle size range exhibited minimal κ_m variation (average $\kappa_m \pm 0.007$),
247 suggesting ideal solution behaviour across the wide RH range (Z. Wang et al., 2017). The
248 average κ_m values (indicated by dashed lines in Figure 3b) of the size-resolved ambient
249 particles varied significantly between 0.016 (<0.056 nm) and 0.44 (1.0 – 1.8 μm) over the
250 investigated RH range. This implies κ_m as a function of size, and indicates distinct chemical
251 compositions for each size fraction. For example, the small increase in κ_m from 0.016 to
252 0.078 for particles <56 nm to 56-100 nm, respectively, is potentially owing to aging
253 processes (Zhang et al., 2023). Aerosol particles <320 nm exhibited very low average κ_m
254 (0.06), which may be primarily due to the strong dominance of organic compounds in these
255 size ranges (Demou et al., 2003; Li et al., 2021; Shi et al., 2022). The particles in the size
256 range of 320 nm – 1 μm were moderately hygroscopic (average $\kappa_m=0.19$) indicating the
257 influence of inorganic salts (Wu et al., 2016). The particles between 1.0 – 1.8 μm exhibited
258 highest κ_m (0.44), which implies the presence of highly hygroscopic material such as sea salt

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

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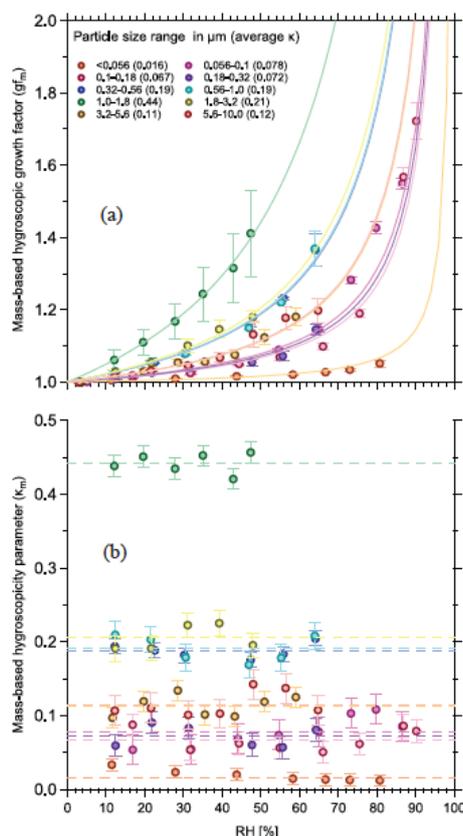
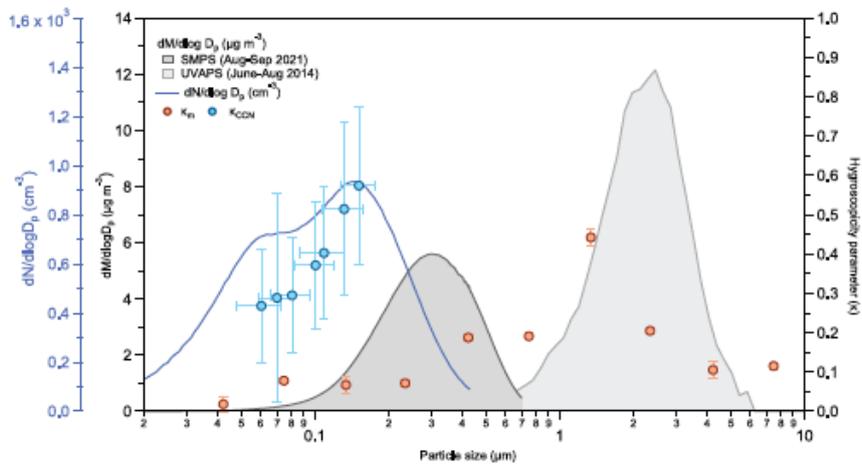


Figure 3. Size-resolved hygroscopicity measurements of ambient aerosol particles at the high-altitude site, Munnar, during the Monsoon season (August-September 2021). (a) Mass-based hygroscopic growth factor (gf_m) derived using a quartz crystal microbalance (QCM) for ten different size ranges of ambient 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.

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

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

324



340 **Figure 4.** Aerosol size distributions, hygroscopicity parameters derived from quartz crystal microbalance
 341 (QCM) experiments (κ_m), and from size-resolved cloud condensation nuclei (CCN) measurements (κ_{CCN}). The
 342 number size distribution obtained using a Scanning Mobility Particle Sizer (SMPS) over the size range of 10 –
 343 430 nm (blue curve) was measured during the sampling period and exhibited a bimodal distribution. The mass
 344 size distributions (dark grey shaded area) were derived based on the aerosol number size distribution by
 345 assuming a density of 1.2 g cm⁻³ for the submicron region. The mass size distribution for the supermicron range
 346 (light grey shaded area) was obtained using Ultraviolet Aerodynamic Particle Sizer (UV-APS) measurements
 347 during the same season (June-August) but for a different year (2014). The hygroscopicity parameters derived
 348 from QCM experiments (κ_m ; orange points) and size-resolved CCN measurements (κ_{CCN} ; blue points) are shown
 349 for the comparison. The error bars for κ_m and κ_{CCN} indicate the measurement uncertainty and variability,
 350 respectively.

351

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

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

366 **4. SUMMARY AND ATMOSPHERIC IMPLICATIONS**

367 We report the first results based on a high-sensitivity QCM technique to investigate the mass-
368 based growth factor and hygroscopicity parameter of size-resolved ambient aerosols over a
369 wide range of RH from a relatively pristine high-altitude site in India. For the investigated
370 size ranges, the pronounced variations in gf_m below the DRH provided an opportunity to
371 better understand the changes in ambient aerosol properties even at low RH values, which
372 may not be revealed by size-based growth factor measurements (Hu et al., 2010; Laskina et
373 al., 2015). These measurements clearly highlight the important and critical role in knowing
374 the changes in aerosol properties based on their RH history for an improved understanding of
375 water uptake, phase transition, and radiative impact of atmospheric aerosol particles (Zhao et
376 al., 2022). Based on the estimated κ_m values, the ambient aerosol particles exhibited the
377 behaviour of an ideal solution (Pajunoja et al., 2015) and strong size-dependent chemical
378 composition. We further observed a pronounced size dependency of the DRH values
379 potentially resulting from complexities of organic solubility in organic-inorganic mixtures
380 (Li et al., 2021). Such a complexity owing to the presence of various organic species may
381 alter the CCN behaviour of atmospheric aerosol particles due to phase transitions and
382 changes in physical properties. The understanding of CCN activation in the supersaturated
383 regime under the assumption of complete solubility of particles is relatively well established.
384 But the understanding of the thermodynamic properties associated with particle growth in the
385 subsaturated regime still remains a challenge. Our findings emphasize the need for additional
386 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.

394 **ASSOCIATED CONTENT**

395

396 **Author contributions**

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

407

408 **Open research**

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

415

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