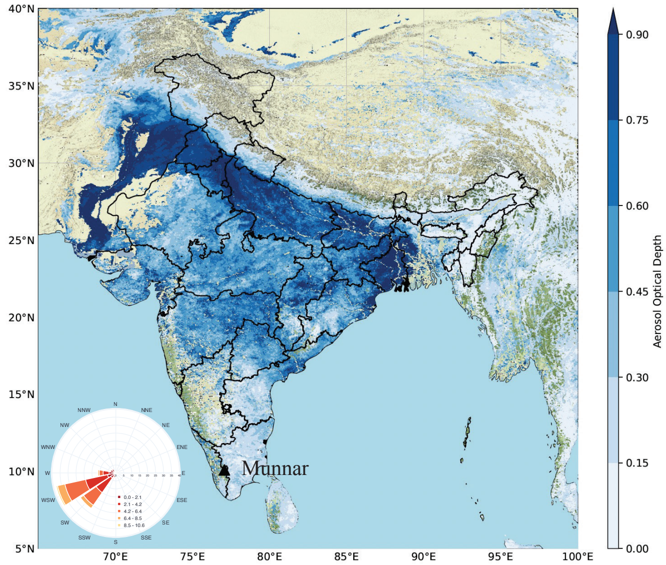

Figure 1. Spatial distribution of average Aerosol Optical Depth (AOD) derived from MODIS (Moderate Resolution Imaging Spectroradiometer) Level 2 data over the Indian continental region during the monsoon season of 2021 (June - September). The AOD distribution clearly indicates a relatively lower aerosol 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 is 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.

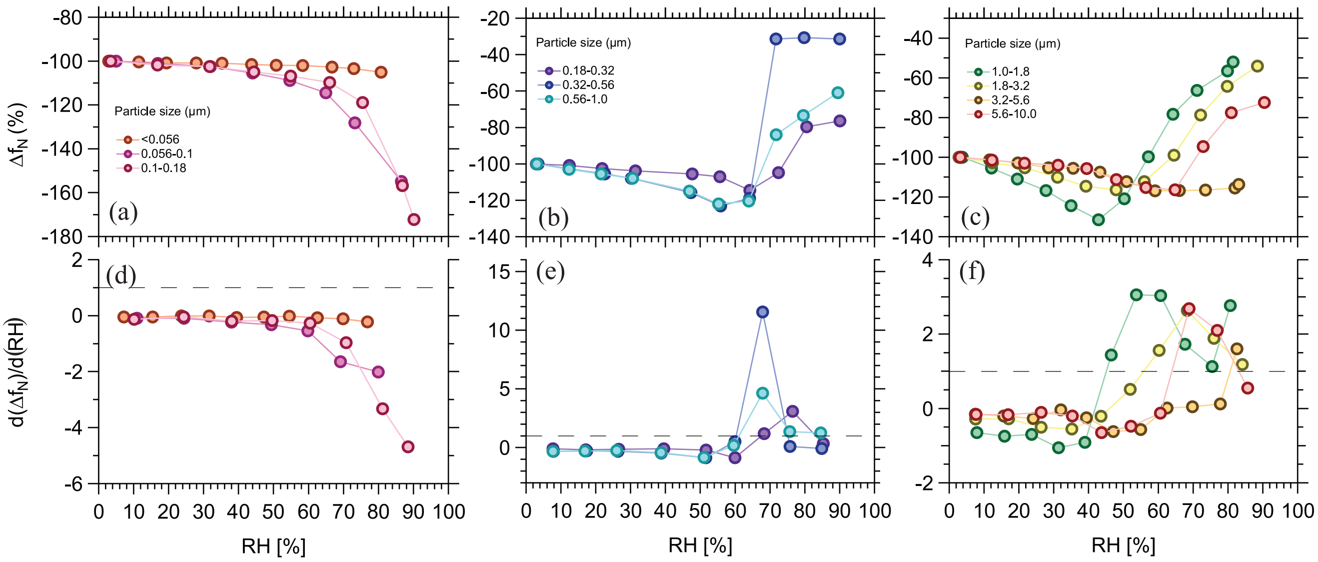

Figure 2. Deliquescence phase transition behaviour of size-resolved ambient aerosol particles from Munnar. For panels a, b, and c, Δf_N represents the change in the oscillation frequency of the quartz crystal microbalance (QCM) sensor resulting due to water uptake by the ambient aerosol particles at different relative humidity (RH) conditions normalised 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.

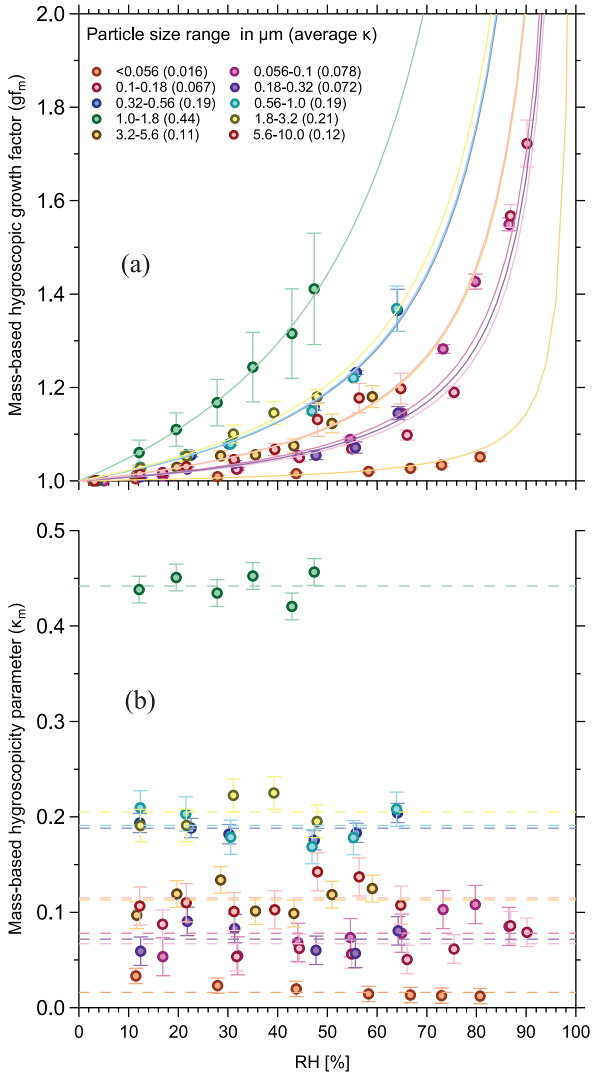

Figure 3. Size-resolved hygroscopicity measurements of ambient aerosol particles at the high-altitude site, Munnar, during the Monsoon season (August-September 2021). (a) Mass-based hygroscopic growth factor (gf_m) derived using a quartz crystal microbalance (QCM) for ten different size ranges of ambient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

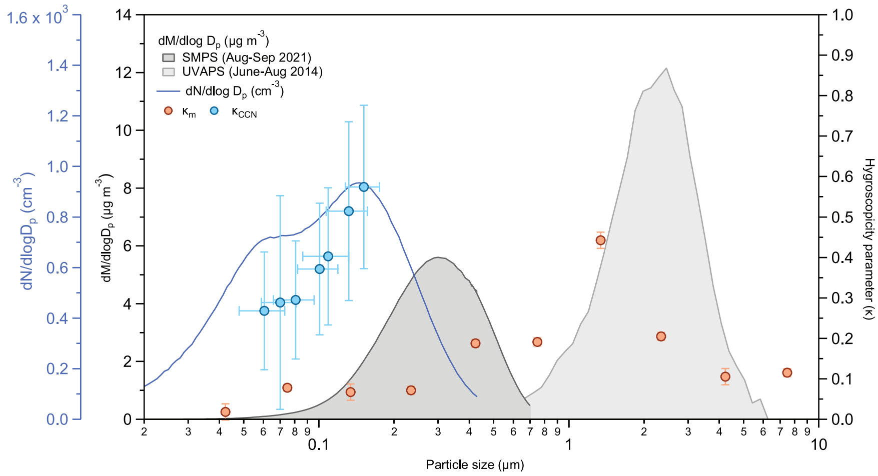

Figure 4. Aerosol size distributions, hygroscopicity parameters derived from quartz crystal microbalance (QCM) experiments (κ_m), and from size-resolved cloud condensation nuclei (CCN) measurements (κ_{CCN}). The number size distribution obtained using a Scanning Mobility Particle Sizer (SMPS) over the 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.