Supporting Information: A Dual-Droplet Approach for Measuring the Hygroscopic Growth of Aqueous Aerosol

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1. Light Scattering Spectrum of a Droplet

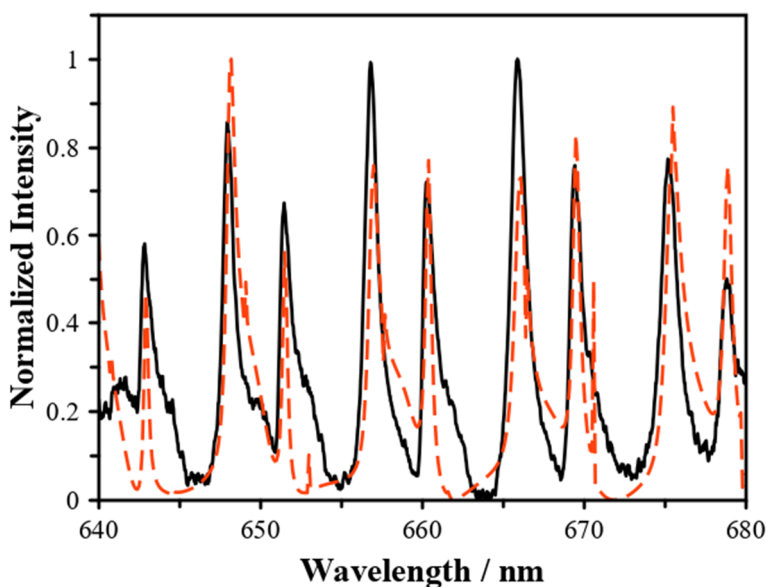



Figure S1: Back-scattered light from a 6000 nm droplet with a refractive index of 1.393 at 589nm. The experimental spectrum (black) is background subtracted and normalized against the illumination intensity profile. A simulated spectrum using Mie theory is shown for comparison (red dash).

2. Deriving a Radial Growth Factor Using Volume-Additive Density

Thermodynamic models such as AIOMFAC report chemical activity and activity coefficients as a function of mass or mole fraction. To convert these to a radial growth factor for comparison to experimental observations, we must assume the density of the solution defined by a particular composition. For binary mixtures explored here, we adopt a volume additive approach, where the radial growth factor (GF) is determined by:

$$GF = \left(\frac{\frac{n_s M_s}{\rho_s} + \frac{n_w M_w}{\rho_w}}{\frac{n_s M_s}{\rho_s}} \right)^{\frac{1}{3}}$$

where the volume of each component is determined from the number of moles (n), the molecular mass (M) and the density of the component (ρ), with w and s subscripts to indicate water and solute, respectively. For inorganic components, the crystal density may be used, although this likely underestimates the effective contribution of the solute to the volume. For organic species, we use the density of the sub-cooled melt predicted using the online tool UManSysProp as the solute density, unless indicated differently in the text. This better reflects the contribution of the solute to the overall volume of the system and yields better agreement to data, as discussed in the main text. The number of moles is derived from the mass or mole fraction output by the thermodynamic model.

3. Measuring the Deliquescent State of NaCl and LiCl

In bulk solution, sodium chloride will precipitate out of solution when the solubility limit is reached, with a water activity of 0.755. For lithium chloride, the water activity is 0.113 at the solubility limit. For both salts in a droplet, a supersaturated solution may form. For the case of NaCl, the concentration will reach up to ~12 m before crystallization occurs, with a water activity around 45%, while for LiCl, crystallization occurs only on prolonged exposure to dry conditions, indicating the water activity is close to zero. When the dry particle is exposed to a gas-phase water activity ($RH/100$) at or above the water activity of the saturated solution, it will take up water and deliquesce to an aqueous solution.

In this work, we use the deliquescence step as a fixed point in the measurement of the RH probe droplet, allowing its dry size to be elucidated and the radial growth factor to be calculated. Figure S2 shows the deliquescence experiment, with the RH increased slowly until water uptake occurs and the dry particle solubilizes to an aqueous solution. When the particle is fully deliquesced, its Mie resonance spectrum becomes resolved and the size is determined. Prior to this, the change in the balancing voltage (V_{DC}) is used as a guide for the onset of deliquescence.

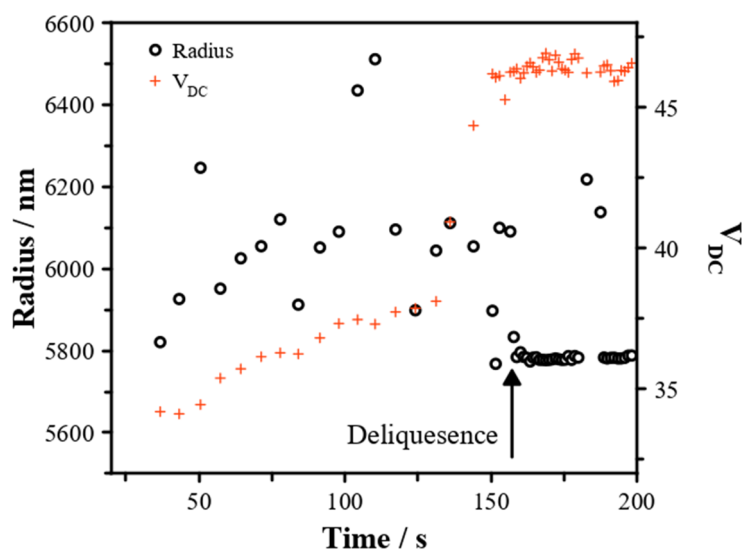
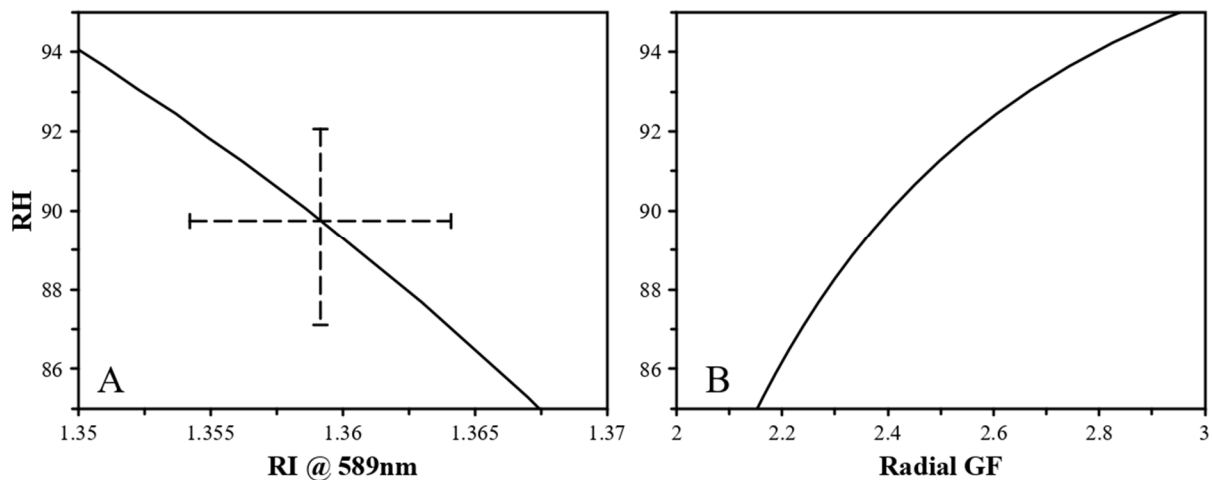



Figure S2: A crystalline NaCl particle was exposed to a slowly increasing RH up to the deliquescence point, at which point it takes up water and becomes an aqueous solution. The radius data is only meaningful for the deliquescence particle, while V_{DC} reflects the aerodynamic radius.

45 **Figure S3:** (A) RH as a function of the RI of NaCl, taken from Cotterell et al. (B) RH as a function of radial GF, derived from E-AIM prediction for NaCl. The uncertainty in the RI is represented by the dashed line, with the corresponding uncertainty in RH shown. The uncertainty in the radial GF is approximated by the line width, translating to an uncertainty in the RH that is ~0.1%.