Supporting Information

Direct Observation and Assessment of Phase States of Ambient and Lab-generated Sub-micron Particles upon Humidification

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Glass transition temperature prediction

A range of glass transition temperature (T_g) for SRFA particles and Soil Organic Particles as a function of RH has been estimated following the methods introduced by [1]:

$$T_g(RH) = \frac{T_{g,w}k_{GT} + f(RH)T_g(RH = 0\%)}{k_{GT} + f(RH)} \tag{S1}$$

where

$$f(RH) = \frac{100 - RH}{RH}\frac{1}{\kappa_{org}}\frac{\rho_{org}}{\rho_w} \tag{S2}$$

where $T_{g,w}$, which is equal to 136 K, is the T_g for pure water, k_{GT} is the Gordon-Taylor constant, κ_{org} is the CCN-derived hygroscopicity parameter of the organic fraction, ρ_{org} and ρ_w are the density of water and organic material, respectively. In this study, a k_{GT} of 1.5 was assumed for both SRFA particles and Soil Organic Particles [1]. T_g(RH = 0%), κ_{org}, and ρ_{org} for SRFA particles and Soil Organic Particles were assumed to be 309 K [1] and 300 K [2], 0.025 [1] and 0.034 [3], and 1.47 g cm^{-3} [1] and 1.9 g cm^{-3} (measured in this study), respectively.

Aspect ratio calculation

In the tilted SEM images, solid particle, which is spherical, will remain the same shape as that at normal stage ((a) in the Figure S1). However, when particle is semisolid and/or liquid, which shows a domelike and/flat shape, respectively, there tilted SEM images will be distorted in the direction of x-axis ((b) in the Figure S1). In this study, we used ImageJ software to measure the maximum particle length in the x-axis and y-axis direction (H and W, respectively). For semisolid and liquid particles, H is the projection of the arc from top to base on the horizontal plane, and W is the projection of base width on the horizontal plane, which is equal to the real particle width. Therefore, to get the real particle height of semisolid and liquid particles collected on the substrate, we did follow corrections:

Figure S2 shows the simplified typical side view of semisolid and liquid particle on a tilted stage (tilted angle = α). BC is the radius of base, which is equal to W/2, AB is the real particle height, and DE is the projection of arc AC, which is equal to H. Thus, ∠DAB=∠CBE=∠α. To calculate real particle height, we have following relationships:

$$BD + BE = H \tag{S1}$$

$$AB = \frac{BD}{\sin \alpha} \tag{S2}$$

$$BE = 0.5W \cos \alpha \tag{S3}$$

Rearrange eq. S1-S3, we get

$$AB = \frac{H}{\sin \alpha} - 0.5W \frac{\cos \alpha}{\sin \alpha} \tag{S4}$$

Therefore, the real aspect ratio (AR') for semisolid and liquid particle is equal to

$$AR^{'} = \frac{W}{AB} = \frac{2AR \sin \alpha}{2 - ARcos\alpha} \tag{S5}$$

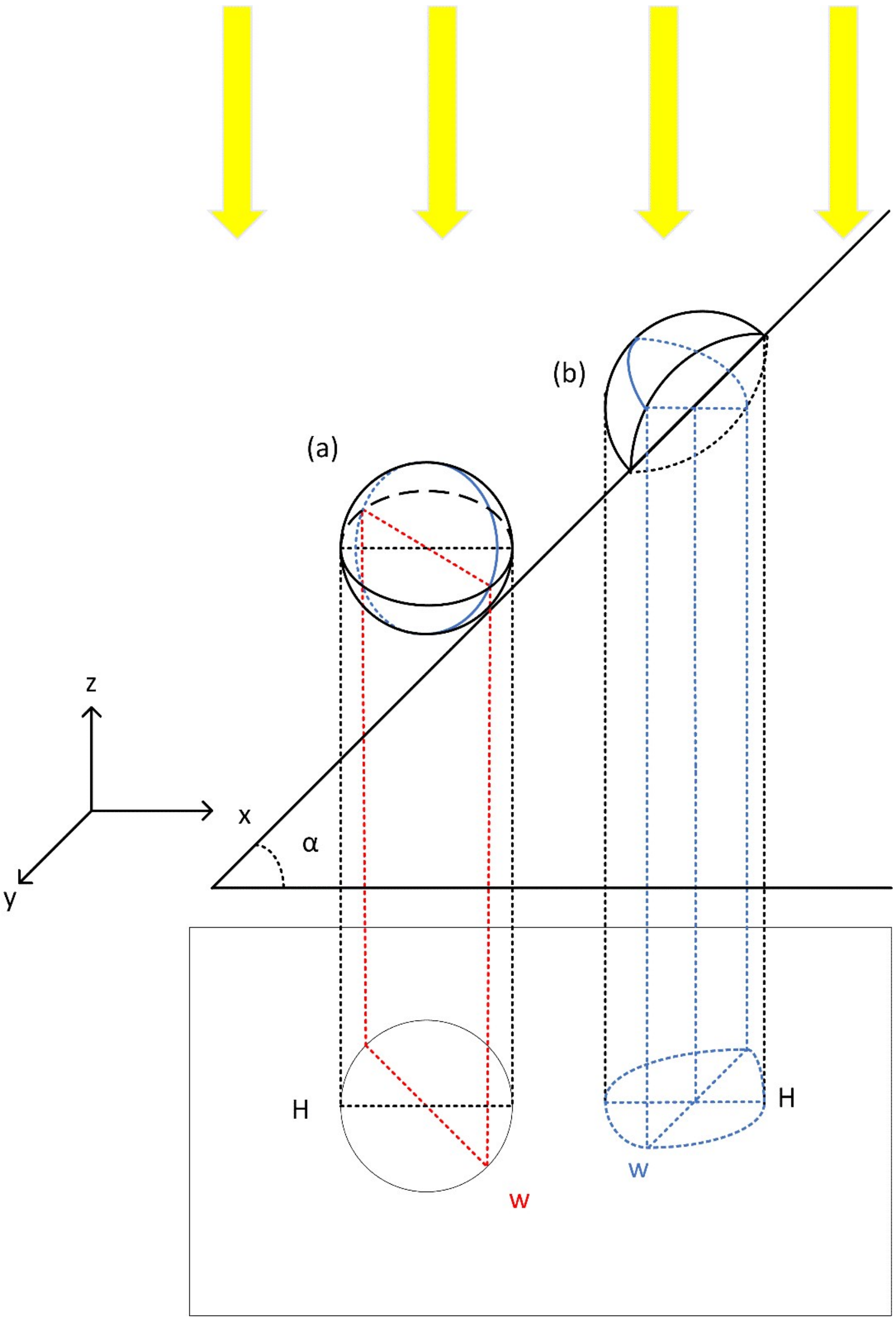

Figure S1: Schematic diagram of the tilted particles and their projection as tilted SEM images. (a) is an example of solid spherical particles, and (b) is an example of semisolid or liquid particles.

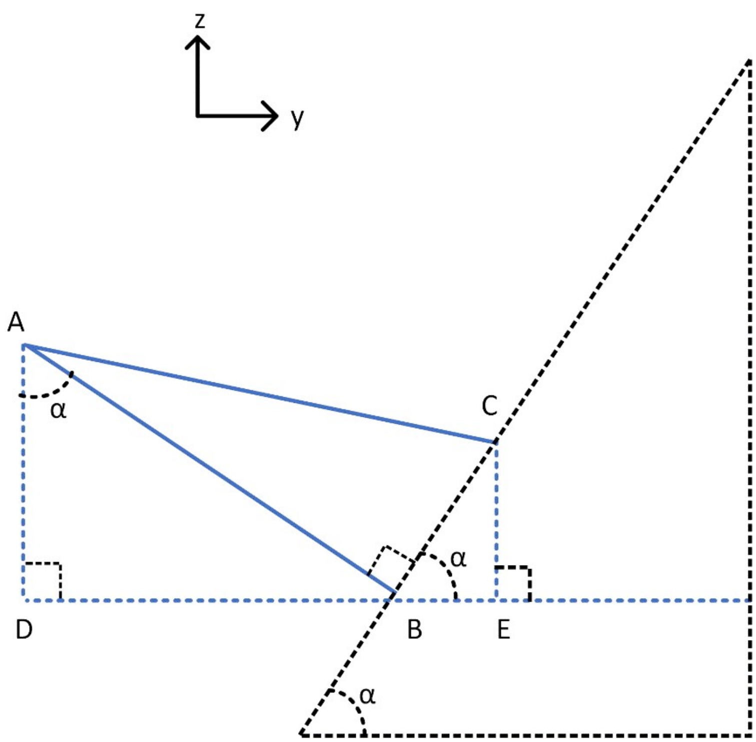

Figure S2: Simplified typical side view of semisolid and liquid particle on a tilted stage (tilted angle = α).

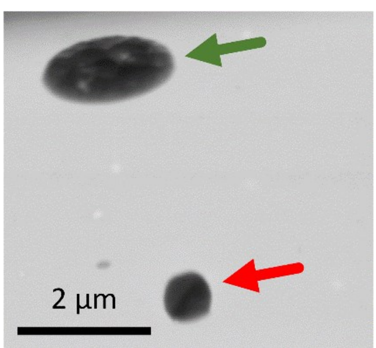

Figure S3. Representative image showing deformation of shape of ambient organic particles when they impact on the substrate. A green arrow indicates a liquid organic particle, and a red arrow indicates a solid organic particle. The liquid ambient organic particle will deform to a flat shape, and the solid ambient organic particle will maintain spherical shape.

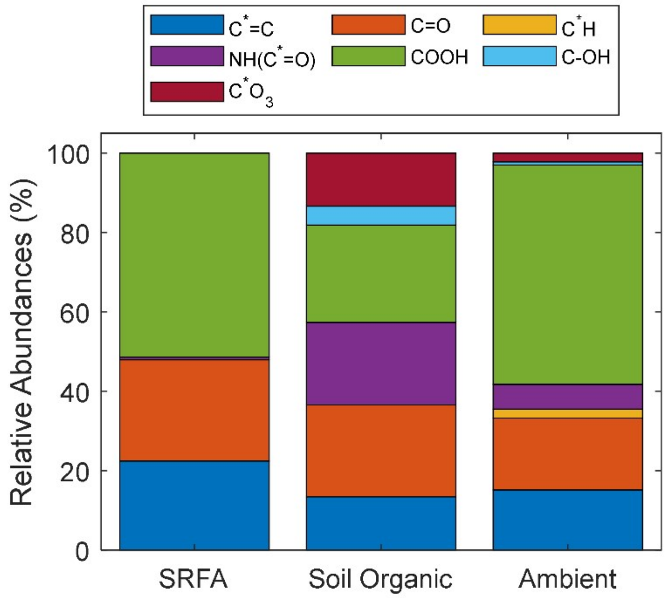

Figure S4: The relative abundances of seven peaks (C=C, C=O, CH, NH(C=O), COOH, C-OH, and CO_3) in SRFA particles, soil organic particles, and Ambient particles.

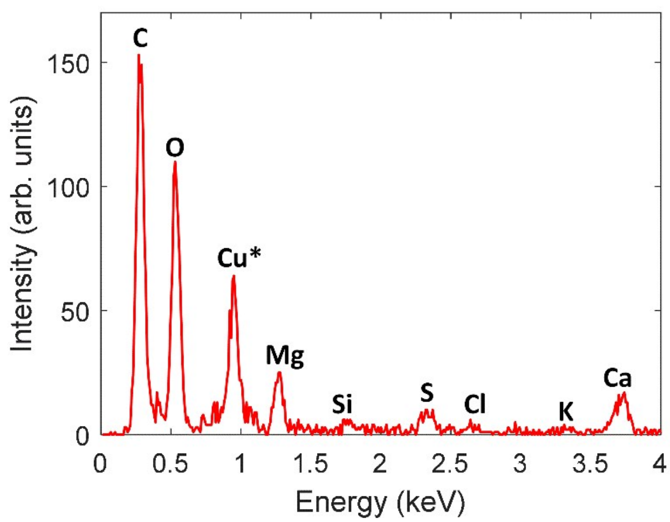

Figure S5: Average particle composition of the soil organic particles sample using CCSEM/EDX analysis. Signal of Cu comes from the Cu TEM grids.

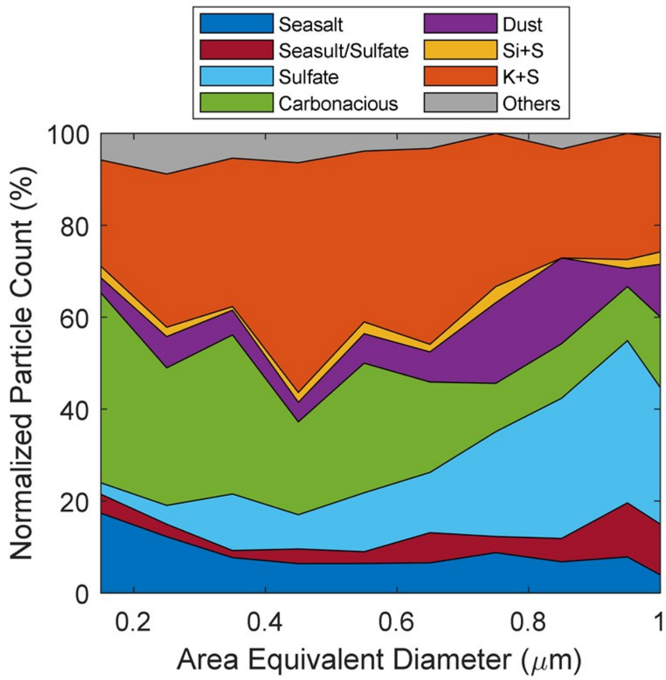

Figure S6: Particle composition of the ambient sample using CCSEM/EDX analysis

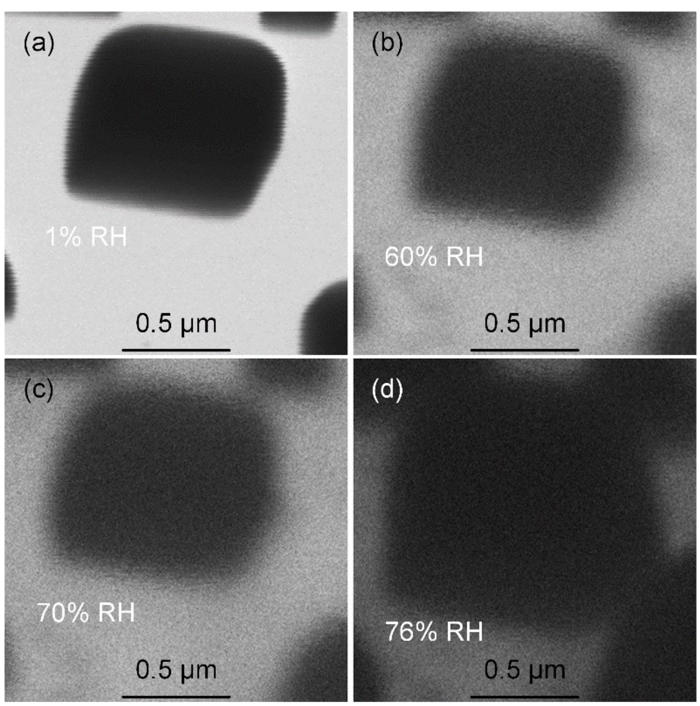

Figure S7: Typical example images of changes in morphology as a result of water uptake by NaCl particles.

References:

(1) Wang, B.; Lambe, A. T.; Massoli, P.; Onasch, T. B.; Davidovits, P.; Worsnop, D. R.; Knopf, D. A. The Deposition Ice Nucleation and Immersion Freezing Potential of Amorphous Secondary Organic Aerosol: Pathways for Ice and Mixed-Phase Cloud Formation. *J. Geophys. Res.* **2012**, *117* (16), 1–12. https://doi.org/10.1029/2012JD018063.

(2) Wang, B.; Harder, T. H.; Kelly, S. T.; Piens, D. S.; China, S.; Kovarik, L.; Keiluweit, M.; Arey, B. W.; Gilles, M. K.; Laskin, A. Airborne Soil Organic Particles Generated by Precipitation. *Nat. Geosci.* **2016**, *9* (6), 433–437. https://doi.org/10.1038/ngeo2705.

(3) Petters, M. D.; Kreidenweis, S. M. A Single Parameter Representation of Hygroscopic Growth and Cloud Condensation Nucleus Activity-Part 3: Including Surfactant Partitioning. *Atmos. Chem. Phys.* **2013**, *13* (2), 1081–1091. https://doi.org/10.5194/acp-13-1081-2013.