SUPPORTING INFORMATION

Synchronized structure and surface tension measurement on individual secondary aerosol particles by low-voltage transmission electron microscopy

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This material contains Supporting Methods, Supporting Tables S1-S2, Supporting Figures S1-S14

Supporting Methods

Details in sample collections

For consistency, these PM samples were taken at the same location in the same time slots in Hung Hom, Hong Kong, from January to July 2019. Each set of samples contained three PM samples, which were taken in three different time slots in a day. The sampling site was the footbridge extended from the main campus of the Hong Kong Polytechnic University in Hung Hom. The other side of the site is the Cross Harbour Tunnel across the Victoria Harbour. Therefore, PMs were able to flow from the sea to the sampling site through a sea breeze. Another characteristic is that the road near this site is heavily trafficked every day, leading to high population of PMs on the near-ground level. To evaluate the changes of PMs in various weather conditions, three PM samples were collected in a day with three time slots: morning (10:00), afternoon (14:00) and evening (18:00). Meteorological factors, including ambient temperature (AT), relative humidity (RH) and visibility, were recorded.

Details in sampling method

Besides the grid, a PM particle counter (DT-9880, CEM) was used for collecting and counting PM levels on the TEM grid. After the TEM grid is mounted at the particle entrance, the particle counter was turned to the side of Cross Harbour Tunnel and started to collect PMs. It took fifteen minutes for collecting each PM sample, to ensure PMs are effectively captured on the grid.

Details in TEM analysis

TEM images under the magnification of 10000X (low mag) demonstrated the overview of samples. High-resolution of the PM (high magnification) images were allowed to investigate the morphology and chemical composition of a single particle in a PM sample. In specific, the JEM-ARM200F transmission electron microscope (TEM) with a CEOS spherical (Cs) aberration corrector was used. The accelerating voltage (60 kV) and exposure time has been minimized to reduce the electron beam damage on the sample. The vacuum value during measurement was better than 1.5×10^{-7} mbar, together with the electron beam current of ca. 10 pA. At the time of performance, the scanning probe size was 1.5 Å. The acquisition time of STEM-HAADF image was 19 μs per pixel in order to minimize electron beam damage. The 512 × 512 pixel images were acquired with the CL aperture of 40 μm and the range of collection angle was 45 to 180 mrad.

Supporting Tables

Table S1. The weather conditions (sampling date/time) for our field campaign in HK.

Temperature	Humidity	Hygroscopic particle contribution(percentage)
16.8°C	72%	72%
17.3°C	56%	86%
19.0°C	60%	79%
19°C	80%	72%
19.8°C	80%	73%
20.0°C	81%	73%
24.5°C	82%	70%
25.7°C	80%	73%
26.0°C	75%	72%

Table S2. The hygroscopic SA contribution (percentage) in the total near-ground PM (for particle sizes >100 nm) under different weather conditions.

Sampling date&time (AT/RH/Visibility)	10:00am	2:00pm	6:30pm
08/01/2019	19.8°C/80%/6km	20.0°C/81%/8km	19°C/80%/6km
23/01/2019	17.3°C/56%/12km	19.0°C/60%/8km	16.8°C/72%/9km
05/03/2019	25.7°C/80%/14km	26.0°C/75%/N.A.	24.5°C/82%/N.A.
26/03/2019	23.2°C/82%/12km	22.5°C/83%/8km	21.6°C/86%/N.A.
18/06/2019	30.0°C/85%/3km	31.5°C/74%/1km	27.5°C/87.5%/1km

Supporting Figures

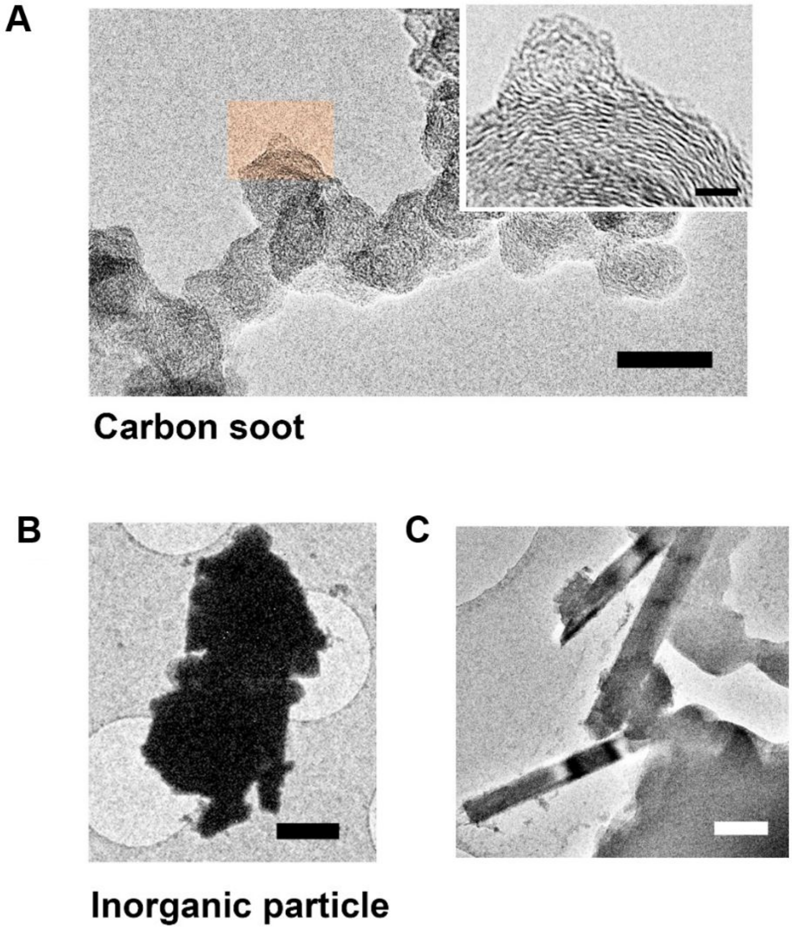

Figure S1. (A) TEM images for typical carbon soot and (B), (C) inorganic particles collected in the atmosphere PM samples in Hung Hom. Scale bars=500 nm.

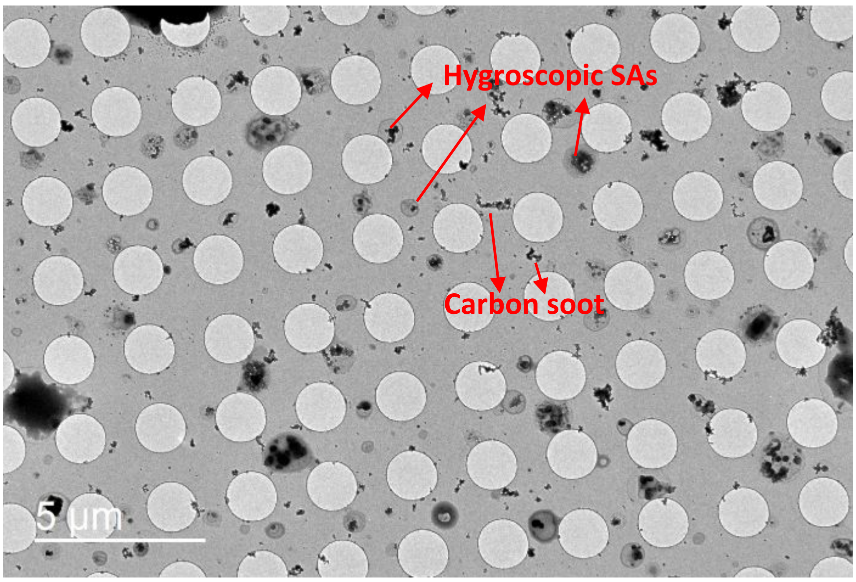

Figure S2. Representative low magnification TEM image for PM samples collected in Hung Hom atmosphere.

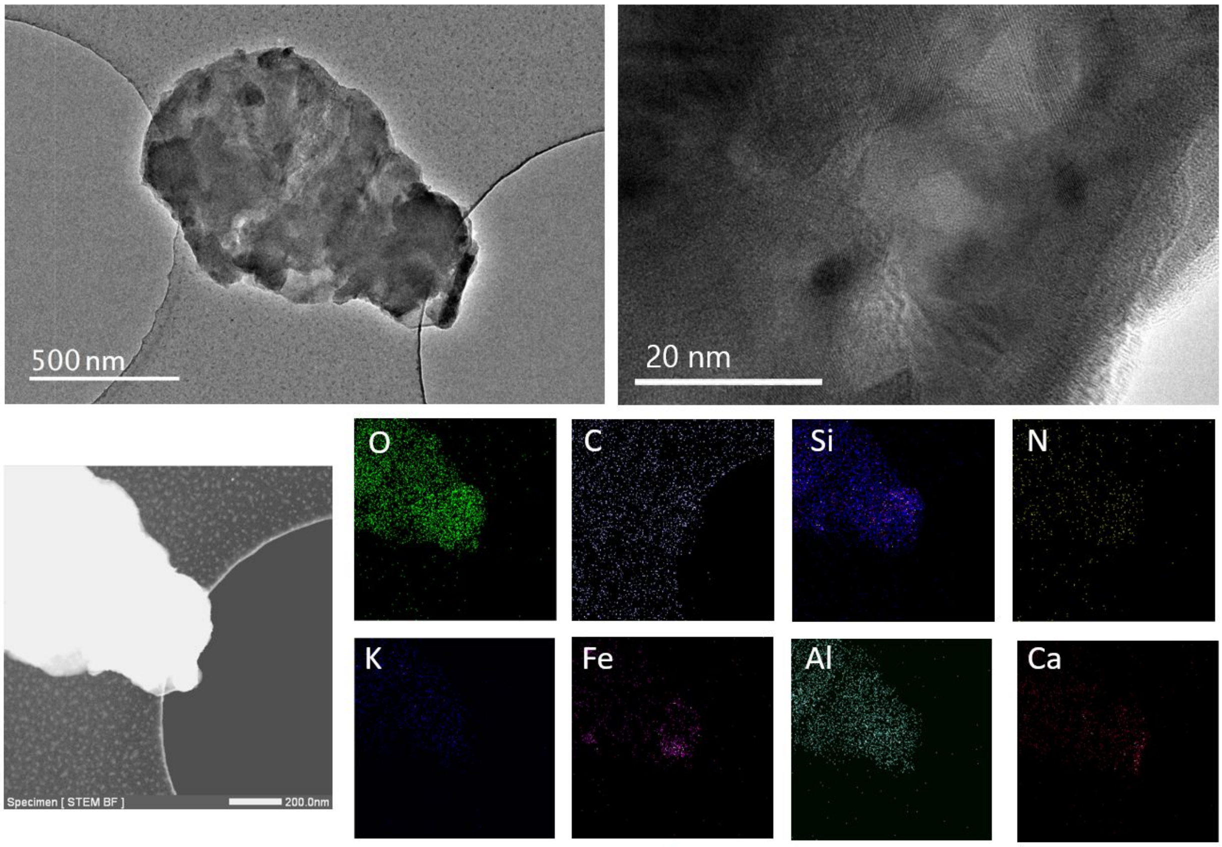

Figure S3. TEM, HAADF and STEM-EDS mapping of particles with metal components from particle collection of SAs.

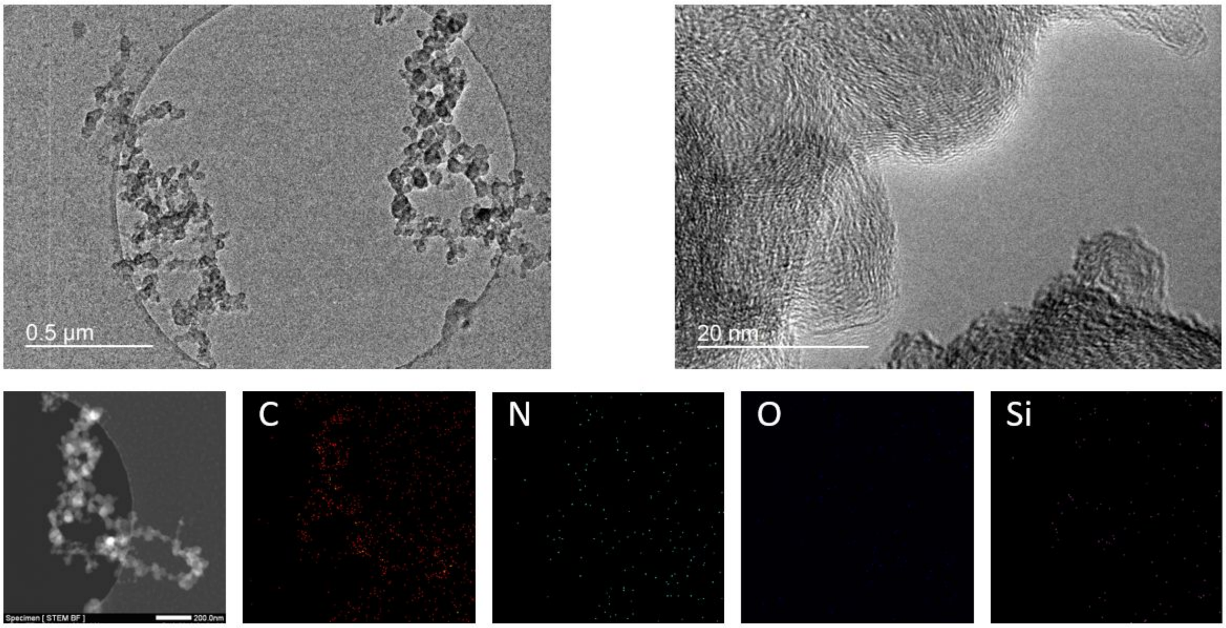

Figure S4. TEM, HAADF and STEM-EDS mapping of onion-shaped carbon particles.

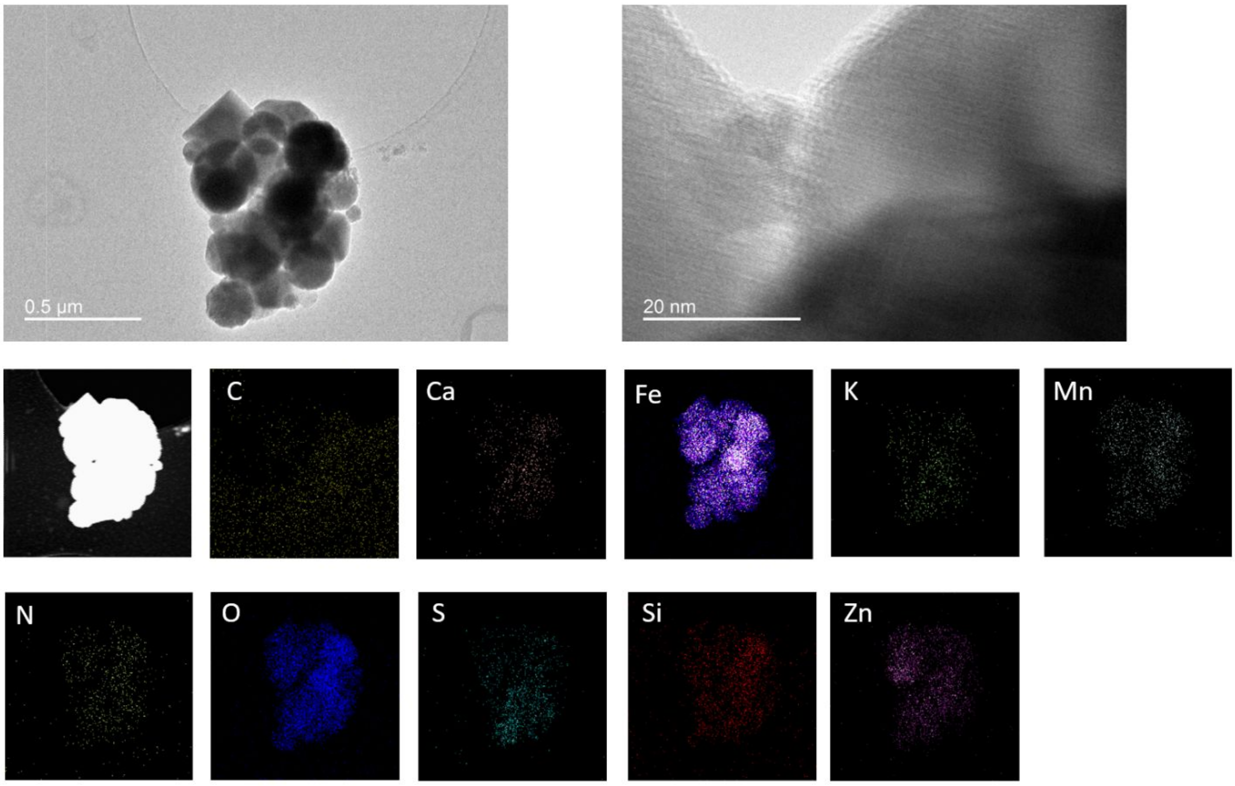

Figure S5. TEM, HAADF and STEM-EDS mapping of cluster particles with metal components from particle collection of SAs.

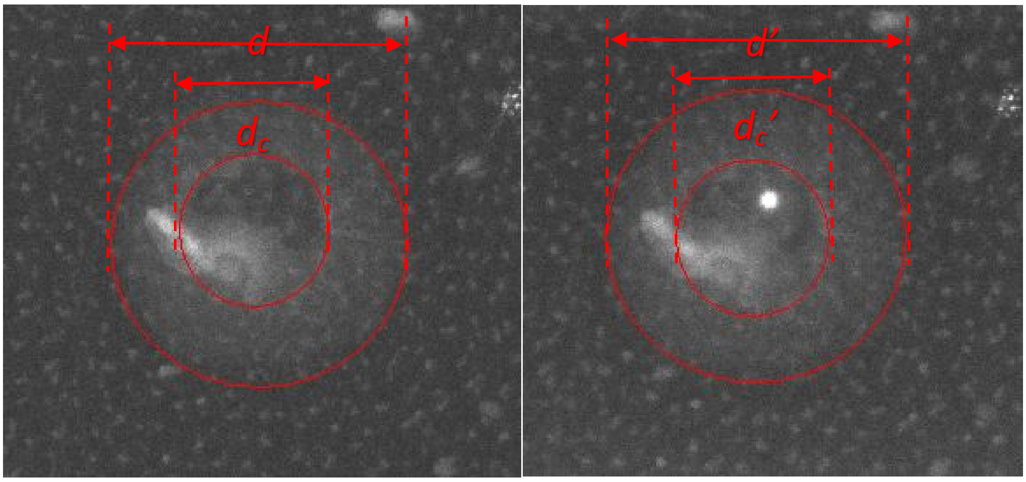

Figure S6. HAADF image of a typical SA before (left) and after (right) STEM-EDS mapping.

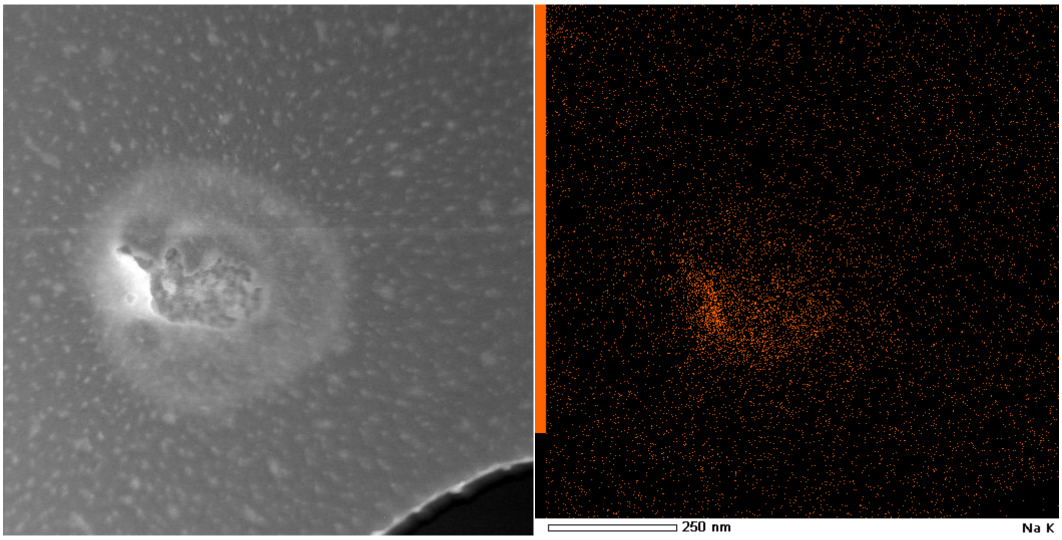

Figure S7. The typical low salt content aerosol particles (STEM image on the left and EDS mapping of Na on the right).

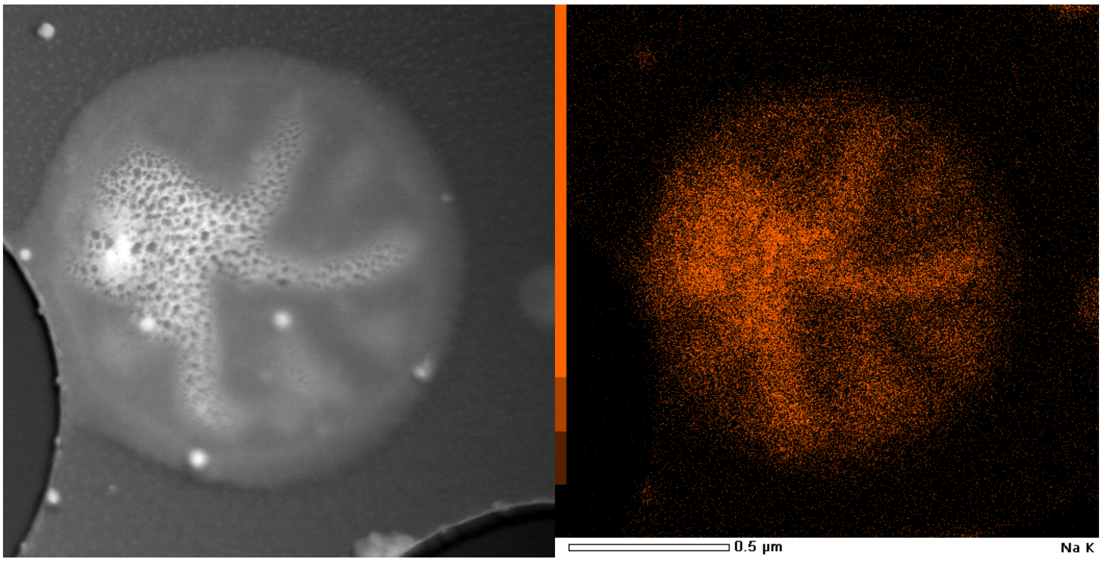

Figure S8. The typical high salt content aerosol particles (STEM image on the left and EDS mapping of Na on the right)

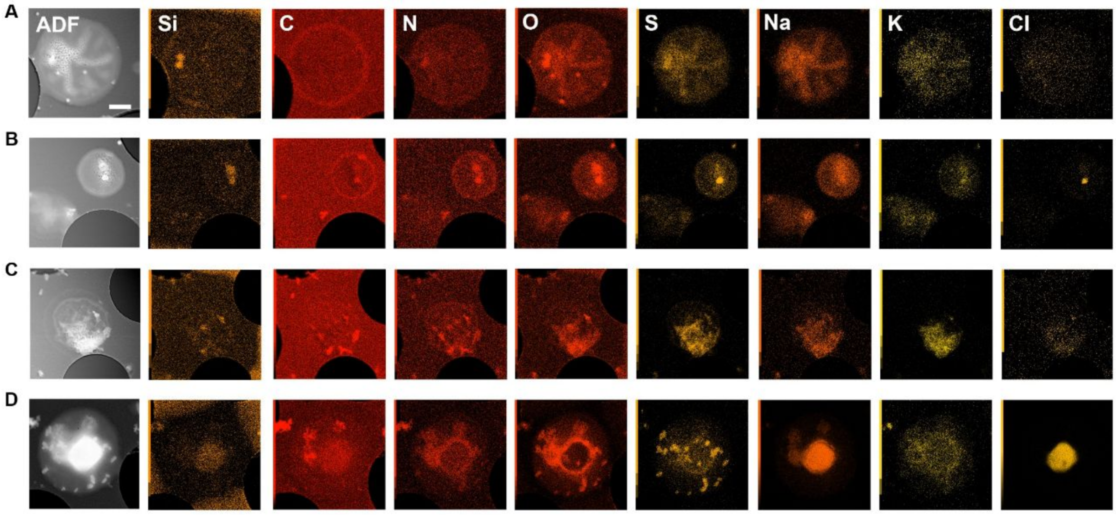

Figure S9. STEM-ADF and EDS mapping of typical SA particles. Scale bar (same for all panel images in this figure) =200 nm.

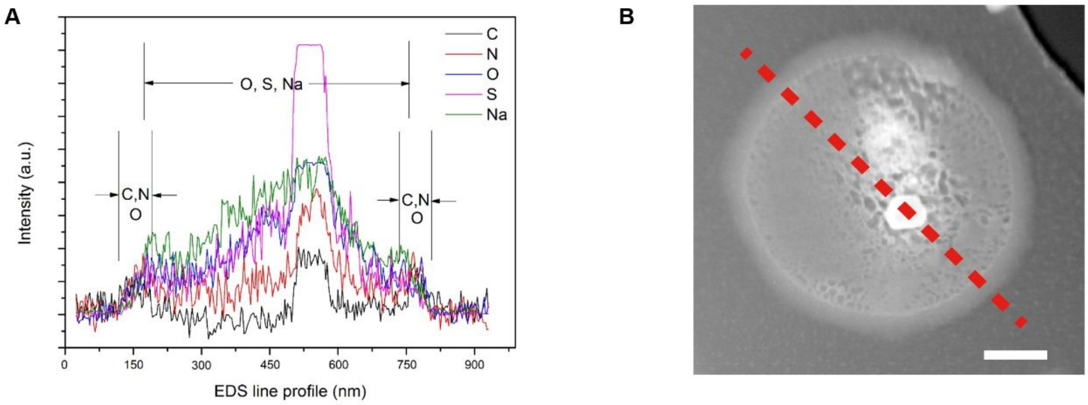

Figure S10. (A) Representative energy dispersive spectra mapping profile for the elements C, N, O, S, Na, corresponding to the red dashed line in HAADF image of SA in (B). Scale bar =100 nm.

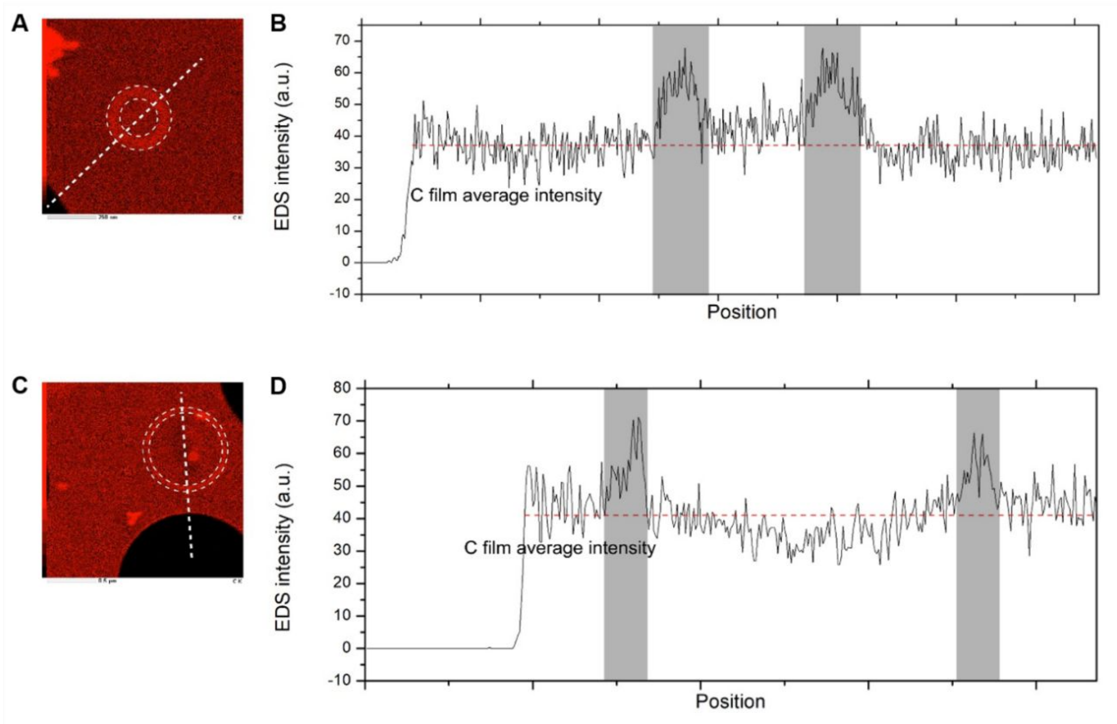

Figure S11. (A), (C) The quantitative EDS mapping for carbon of PM particles. The dashed circles highlight the integrated signal area of carbon content of the shells of PM particles. (B), (D) The extracted EDS signals along the white straight dashed lines in (A), (C) respectively. The signals for supporting carbon film area are used for normalization and error analysis. Red dashed lines show the averaged C film EDS intensity. Gray zones correspond to the shells of PM particles.

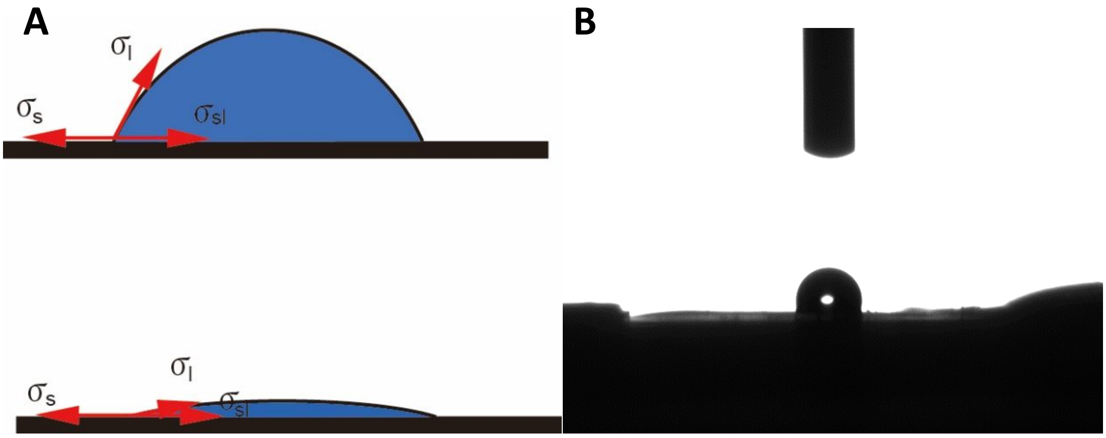

Figure S12. A. The illustration of evaporation of SA particle on carbon film. **B**. The in-house surface tension measurement of contact angle between pure water (zero surface tension reduction) and amorphous carbon film which we used to collect the SA particles in the field study. The droplet volume is 0.2μL.

The absolute values of surface tension reduction are about the dimensionless surface tension reduction results. The (-1.0) of the dimensionless surface tension reduction corresponds to the maximum surface tension reduction which is completely wetting (contact angle close to 0^{o}) between SA particle and the carbon film, in approx. 200 nm diameter aerosol particles which do not have the dome-lowering evaporation mode, thus a 24% reduction of surface tension compared to pure water can be derived from the contact angle measurement result between the amorphous carbon film (of TEM grid) and pure water provided by our in-house contact angle measurements, see above Figure S12. By using the contact angle result and the surface tension measurement results in field studies, the surface tension of the SA particles in our field campaign should be within the range of 54~72 mN/m.

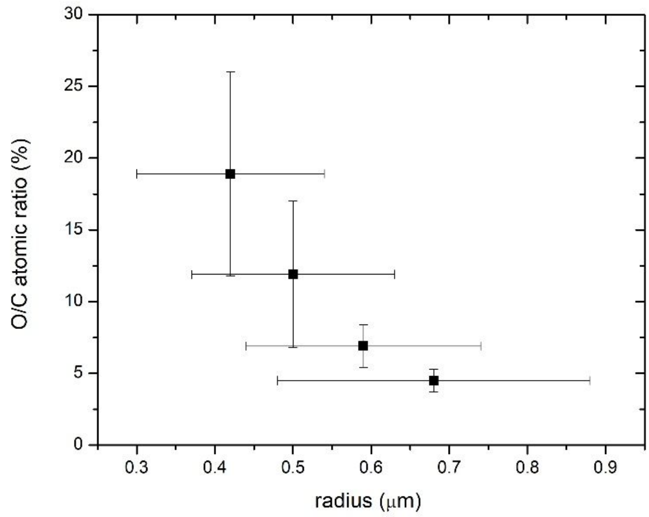

Figure S13. The mean O/C atomic ratio in shell versus mean radius of hygroscopic SA particles, measured under different weather conditions.

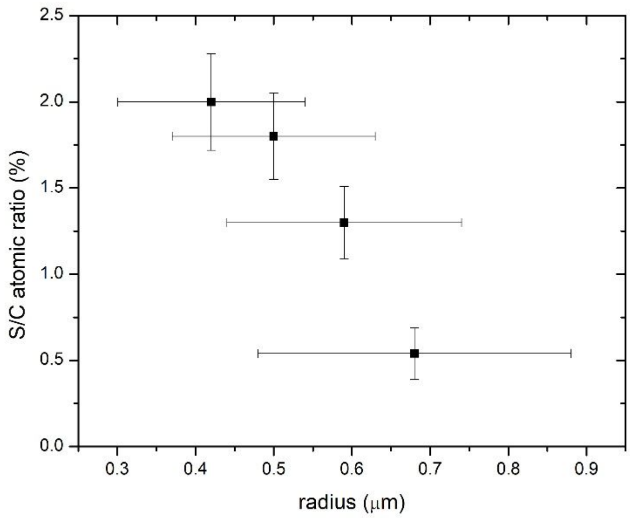

Figure S14. The mean S/C atomic ratio in shell versus mean radius of hygroscopic SA particles, measured under different weather conditions.