



Supplement of

Single particle characterization using the soot particle aerosol mass spectrometer (SP-AMS)

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Tables:

| m/z | frag_organic | frag_rBC [#] | rBC fragments |
|-----|----------------------|-----------------------|---------------|
| 12 | 12,-frag_rBC[12] | 0.61*frag_rBC[36] | C_1^+ |
| 24 | | 24,-frag_sulfate[24] | C_2^+ |
| 36 | | 36,-frag_air[36] | C_3^+ |
| 48 | 0.5*frag_organic[62] | 0.057*frag_rBC[36] | C_4^+ |
| 60 | 60,-frag_rBC[60] | 0.047*frag_rBC[36] | ${\rm C_5}^+$ |

Table S1: The refractory black carbon (rBC) column of the fragmentation table in Squirrel.

[#] The C_1/C_3 ratio of ambient rBC was determined using the C_1/C_3 ratio of Regal Black, and the C_4/C_3 and C_5/C_3 ratios of ambient rBC were determined using the high resolution fitting data of ambient measurements.

Table S2: The simplified fragmentation patterns of each aerosol species for the calculation of chemical compositions for individual single particles from UMR cluster results.

| Aerosol species | The simplified fragmentation patterns |
|-----------------|--|
| Total | All ions – m/z 14 (N ⁺) – m/z 15 (NH ⁺) – m/z 16 (NH ₂ ⁺) – m/z 17 (NH ₃ ⁺) – m/z 18 (H ₂ O ⁺) – m/z 28 (N ₂ ⁺) – m/z 32 (O ₂ ⁺) – m/z 39 (K ⁺) |
| Nitrate | $m/z \ 30 \ (NO^+) + m/z \ 46 \ (NO_2^+)$ |
| Sulfate | $(m/z \ 48 \ - \ 0.047 \ m/z \ 36) \ (SO^{+}) + m/z \ 64 \ (SO_{2}^{+}) + 0.203 \ m/z \ 64$ $(HSO_{2}^{+}) + m/z \ 80 \ (SO_{3}^{+}) + 0.926 \ m/z \ 80 \ (HSO_{3}^{+}) + m/z \ 98 \ (H_{2}SO_{4}^{+})$ |
| rBC | $0.61*m/z \ 36 \ (C^+) + m/z \ 24 \ (C_2^+) + m/z \ 36 \ (C_3^+) + 0.057*m/z \ 36 \ (C_4^+) + 0.047*m/z \ 36 \ (C_5^+)$ |
| Organics | Total – Nitrate – Sulfate – rBC |

Figures:



Figure S1: The size distributions (from light scattering signal, d_{va}) of prompt particles in terms of (a and c) the total light scattering signal intensity and (b and d) the total number of ions in an individual particle. The green dots represent the prompt particles.



Figure S2: The total distance between the cluster centers and each single particle data obtained from k-means clustering analysis. The dashed line indicates the number of clusters (12) for the interpretation of the mixing state of ambient particles in the current study.



Figure S3: Van-Krevelen diagram: The grey area represents typical values for ambient organic aerosol measured in North Hemisphere (Ng et al. 2010). HOA, COA and OOA coordinates were obtained by PMF analysis of the organic components using the laser off measurements.

Reference:

Ng, N. L., Canagaratna, M. R., Zhang, Q., Jimenez, J. L., Tian, J., Ulbrich, I. M., Kroll, J. H., Docherty, K. S., Chhabra, P. S., Bahreini, R., Murphy, S. M., Seinfeld, J. H., Hildebrandt, L., Donahue, N. M., DeCarlo, P. F., Lanz, V. A., Prevot, A. S. H., Dinar, E., Rudich, Y. and Worsnop, D. R.: Organic aerosol components observed in Northern Hemispheric datasets from Aerosol Mass Spectrometry, Atmospheric Chemistry and Physics, 10, 4625-4641, 2010.



Figure S4: Mass fraction contributions of each PMF factor (Blue: HOA, Red: COA, Orange: OOA, Grey: Residual) to the total organics from IR laser off measurement.



Figure S5: The time series of organic factors obtained from the IR laser on measurements without (a-c) and including rBC signals (d-f) in the PMF analysis.



Figure S6: Normalized mass spectra of PMF organic factors from laser on mode measurement (without rBC signals in the PMF analysis): (a) HOA factor, (b) COA factor, (c) OOA factor.



Figure S7: Comparison between the IR laser on and off measurements: (a) NH_4^+ (b) SO_4^{2-} (c) NO_3^- and (d) Organic. Dashed line = 1:1 line, red line = data fitting.



Figure S8: Comparison between PMF results from the IR laser on and off measurements: (a) HOA (b) COA and (c) OOA. Dashed line = 1:1 line, red line = data fitting. (d) Difference between the two operation modes as a function of rBC fraction. Dashed line = data fitting.



Figure S9: (a) Size distributions of particle number from the ambient single particle measurements (Red: laser on mode, Blue: laser off mode, Green: difference between laser on and laser off modes). The grey dashed line represents the counting efficiency (the ratio of light scattering counts and condensation particle counter signals (LS/CPC), left axis) of the light scattering module for ammonium nitrate particles. The purple dashed line indicates an estimated light scattering cut-off d_{va} for spherical hydrocarbon oil droplets (i.e., shape factor = 1 and density = 1 g/cm³). (b) Ratio of particle numbers (Green circle) and total ion signals (Purple square) between the laser on and laser off modes.



Figure S10: The number of particles (white circles) and the ion fractions of nitrate (blue), sulfate (red), organics (green) and rBC (black) in the 12 preliminary clusters obtained from the k-means clustering analysis.

Nitrate cluster (Figure 8a) = Clusters 1, 6, 8, and 11

Sulfate cluster (Figure 8b) = Cluster 7

HOA cluster (Figure 8c) = Cluster 5

rBC cluster (Figure 8d) = Cluster 9

COA cluster (Figure 8e) = Clusters 2 and 4

OOA cluster1 (Figure 8f) = Cluster 3 and 12

OOA cluster2 (Figure 8g) = Cluster 10



Figure S11: Four preliminary nitrate-rich clusters are combined into a nitrate-rich final cluster, i.e., shown in Figure 8a, h, and p.