Parametrizations of size distribution and refractive index of biomass burning organic aerosol with black carbon content

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1. Methods

1.1 The modelling method of $AAE_{BC,\lambda-880}$

The absorption coefficient $\sigma_{abs,Mie,\lambda}$ was obtained by the following:

$$\sigma_{abs,Mie,\lambda} = \int Q_{abs,Mie,\lambda}(Dp) * \left(\frac{\pi}{4} D_p^2\right) N\left(logD_p\right) * d_{logD_p} \quad (1)$$

where $N\left(logD_p\right)$ is the PNSD function, $Q_{abs,Mie,\lambda}$ is absorption efficiency which is simulated using the Mie-theory, D_p is the particle. For D_p bin, particles are classified into three types: non-light-absorbing particles (Non-BC), BC-containing particles and pure BC particles. Parameters r_{ext} defined as mass fraction of pure externally mixed BC (M_{ext}) to total BC mass (M_{BC}) in different diameter bins and R_NBC defined as the number fraction of particles that does not contain BC are used to represent BC mixing state.The PNSD of Non-BC particles ($N(logD_p)_{Non-BC}$) and BC-containing particles ($N(logD_p)_{BC}$) can be given by the following equations:

$$N(logD_p)_{Non-BC} = N(logD_p)_{measure} * R\_NBC \quad (2)$$

$$N(logD_p)_{BC} = N(logD_p)_{measure} * (1 - R\_NBC) \quad (3)$$

$N(logD_p)_{measure}$ is the PNSD measured by the SMPS and APS. The PNSD of pure BC particles ($N(logD_p)_{ext}$) and core–shell mixed BC particles ($N(logD_p)_{core-shell}$) can be given by the following equations:

$$r_{ext} = \frac{M_{ext}}{M_{BC}} \quad (4)$$

$$N(logD_p)_{ext} = N(logD_p)_{BC} * f_{ext}(Dp) \quad (5)$$

$$N(logD_p)_{core-shell} = N(logD_p)_{BC} * (1 - f_{ext}(Dp) \quad (6)$$

$f_{ext}(Dp)$ is volume fraction of pure BC at each diameter bin which can be calculated by :

$$f_{ext}(Dp) = \frac{M_{BC}(Dp) * r_{ext}}{\rho_{BC} * N(logD_p)_{BC} * (\frac{\pi}{6} D_p^3)} \text{[1]} \quad (7)$$

where ρ_{BC} is the density of BC (1.5 g cm^{-3}[2]); M_{BC} is derived from AE33. Size distribution of BC mass $M_{BC}(Dp)$ is calculated based on the normalized Cx fraction $f_{Cx}(Dp)$ at each diameter bin:

$$M_{BC}(Dp) = M_{BC} \times f_{Cx}(Dp) \quad (8)$$

In addition to the PNSD of the three types of particles mentioned above, another key parameter of the core-shell model is the diameter of the BC core, at each diameter bin the D_{core} is calculated as:

$$D_{core} = \left(\frac{6\times M_{BC}(Dp)*(1-r_{ext})}{\rho_{BC}*\pi}\right)^{1/3} \quad (9)$$

After obtaining the absorption coefficient at each wavelength, the AAE at two wavelengths is calculated as the following equation:

$$\mathrm{AAE}_{\lambda 1-\lambda 2} = \frac{\ln(\sigma_{\mathrm{abs},\lambda 1})-\ln(\sigma_{\mathrm{abs},\lambda 2})}{\ln(\lambda_1)-\ln(\lambda_2)} \quad (10)$$

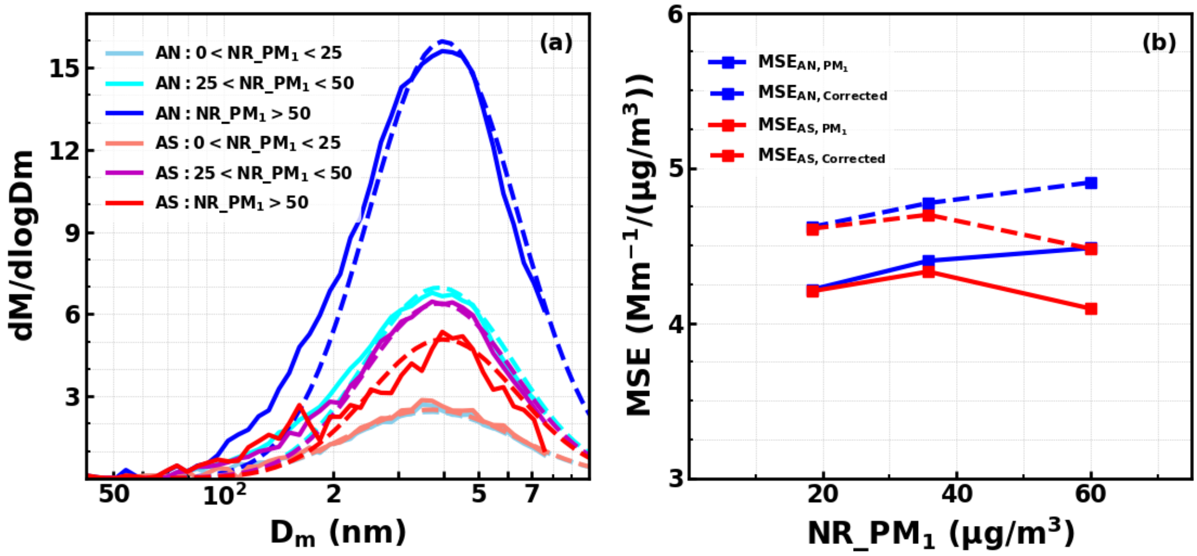

Figure S1. (a) Average size distributions of AS and AN under different NR_PM1 conditions, dashed lines are lognormal fitting curves; (b) Simulated PM1 MSE of AS and AN under different NR_PM1 conditions and corrected ones

1.2 Mass Scattering Efficiency calculations for different aerosol components.

The size distributions of ammonium (AN) nitrate and ammonium sulfate (AS) paired from measured size distributions of nitrate, ammonium and sulfate by the SP-AMS under different non-refractory mass concentrations of PM_1 (NR_PM_1, PM_1 corresponds to aerosols with aerodynamic diameter less than 1 μm) is shown in Fig.S1a. Note that the D_m converted by assuming an aerosol density of 1.6 g/cm^3 from the SP-AMS vacuum aerodynamic diameter Dva. The $MSE_{AN,PM1}$ defined as $MSE_{PM1} = \frac{\sigma_{sp,PM1}}{AN_{PM1}}$ under different NR_PM_1 conditions and results of $MSE_{AS,PM1}$ are shown in Fig.S1b. The density of AS and AN are 1.769 and 1.72 g/cm^3, and used refractive index of AS and AN is 1.53-10^{-7}i. However, in this study, the aerosol scattering coefficient of PM_{10} (aerosols with aerodynamic diameter less than 10 μm) and the aerosol mass concentration of PM_1 is measured by the SP-AMS. Therefore, the corrected MSE defined as $MSE_{Corrected} = \frac{\sigma_{sp,PM10}}{AN_{PM1}} = MSE_{PM1} \times C$ is simulated and also shown in Fig.1b, and C is the correction factor. The average $MSE_{Corrected}$ values for AS and AN are 4.6 and 4.8 m^2/g, and used in this study. The MSE_{HOA} is simulated on the basis of identified average HOA D_{gv} and σ_g of 180 nm and 1.46 using the Mie theory in combination with calculated HOA density of 1.15 g/cm3. The calculated MSE_{HOA} is 3.2 m^2/g. The MSE_{BC} is simulated using the normalized Cx distribution shape shown in Fig.s4, and assuming an BC density of 1.5 g/cm3 and BC refractive index of 1.8-0.54i. The calculated MSE_{BC} is 2.8 m^2/g. The average size distributions of OA measured by the SP-AMS during the entire campaign is shown in Fig.S2a. The average size distributions of HOA and BBOA calculated from average HOA and BBOA mass concentrations together with identified average D_{gv} and σ_g of HOA and BBOA identified from SP-AMS measurements (175, 395 and 1.46,

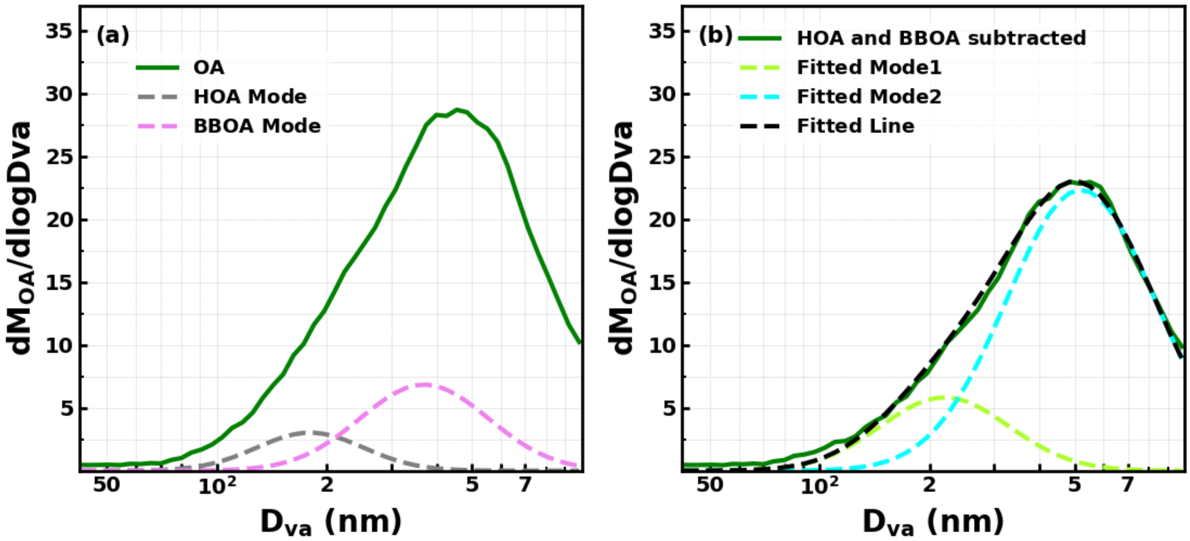

Figure S2. **(a)** Average OA size distributions, and parts associated with HOA and BBOA, Dva is the vacuum aerodynamic diameter. **(b)** The average OA size distributions with contributions of HOA and BBOA subtracted, and the remaining can be well fitted using two lognormal modes.

1.55 for HOA and BBOA as shown in Fig.Sx) are also shown in Fig.S2a. If the contributions of HOA and BBOA are subtracted (Fig.S2b), the OA size distribution can be well fitted using two lognormal modes. The mass concentration of fitted Mode 1 is consistent that of aBBOA and obviously different with mass concentrations of other remaining OA factors, and mass concentration of fitted Mode 2 is consistent with the sum of LOOA, MOOA and Night-OA. Thus, the Mode 1 is identified as the size distribution of aBBOA. The Mode 2 of LOOA， MOOA and Night-OA suggests that most secondary organic aerosols during this campaign are likely internally mixed. Thus the

MSE_{aBBOA} is simulated as 4.5 m^2/g with identified size distribution of Mode 1 and calculated aBBOA density g/cm3 using the scheme proposed by [3]. Corrected MSE of MOOA, Night-OA, LOOA (MSE_{SOA}) is calculated as 6.3 m^2/g with identified size distribution of Mode 2 and estimated average density of 1.31 g/cm3 using volume weighting rule.

1.3 Refractive index retrieval

Determined BBOA MSEs and MAEs were converted to Volume scattering or absorption efficiency (VSE or VAE) through VSE=MSE*density. With given geometric mean (Dgn) and standard deviation (σg) values of the PNSD. The assuming total number concentration of 1000 (Ntot=1000 /cm3), PNSD can be given as :

$$N(logDp) = \frac{dN}{dlogDp}(Dp) = \frac{\mathrm{Ntot}}{\sqrt{2\pi}\log(\sigma_g)}\exp\left[-\frac{(\log(D_p)-\log(D_{gn}))^2}{2\log^2\sigma_g}\right] \quad (11)$$

The with given refractive index of $m = m_{R,BBOA} + m_{i,BBOA} \times i$, the aerosol scattering efficiency Q_{sca} and absorption efficiency Q_{abs} can be calculated using the Mie theory. And then scattering and absorption coefficients of bulk aerosols can be derived as:

$$\sigma_{sca}(\lambda) = \int_0^{D_p^{max}} Q_{sca}(m,\lambda,D_p) \times \frac{\pi}{4} D_p^2 \times N(logD_p) \times dlogD_p \quad (12)$$

$$\sigma_{abs}(\lambda) = \int_0^{D_p^{max}} Q_{abs}(m,\lambda,D_p) \times \frac{\pi}{4} D_p^2 \times N(logD_p) \times dlogD_p \quad (13)$$

Where λ is the optical wavelength, and D_p^{max} of 2500 nm is set. The total volume concentration can be calculated as:

$$Vtot = \int_0^{D_p^{max}} \frac{\pi}{6} D_p^3 \times N(logD_p) \times dlogD_p \quad (14)$$

Then the VSE and MAE can be calculated as:

$$VSE(\lambda) = \frac{\sigma_{sca}(\lambda)}{Vtot} \quad (15)$$

$$VAE(\lambda) = \frac{\sigma_{abs}(\lambda)}{Vtot} \quad (16)$$

The $m_{R,BBOA}$ was retrieved through varying $m_{R,BBOA}$ in iterations to find a $m_{R,BBOA}$ with which the derived VSE can be reproduced, in these iterations $m_{i,BBOA}$ was parameterized with the corresponding ΔCO/ΔBBOA using the relationships determined in Sect.3.4. The $m_{i,BBOA}$ was retrieved through varying $m_{i,BBOA}$ in iterations to find a $m_{R,BBOA}$ with which the derived VAE can be reproduced, and $m_{R,BBOA}$ are fixed as 1.6 due to that sensitivity tests show that very small influences of $m_{R,BBOA}$ variations on $m_{i,BBOA}$

retrieval.

2. Other figures

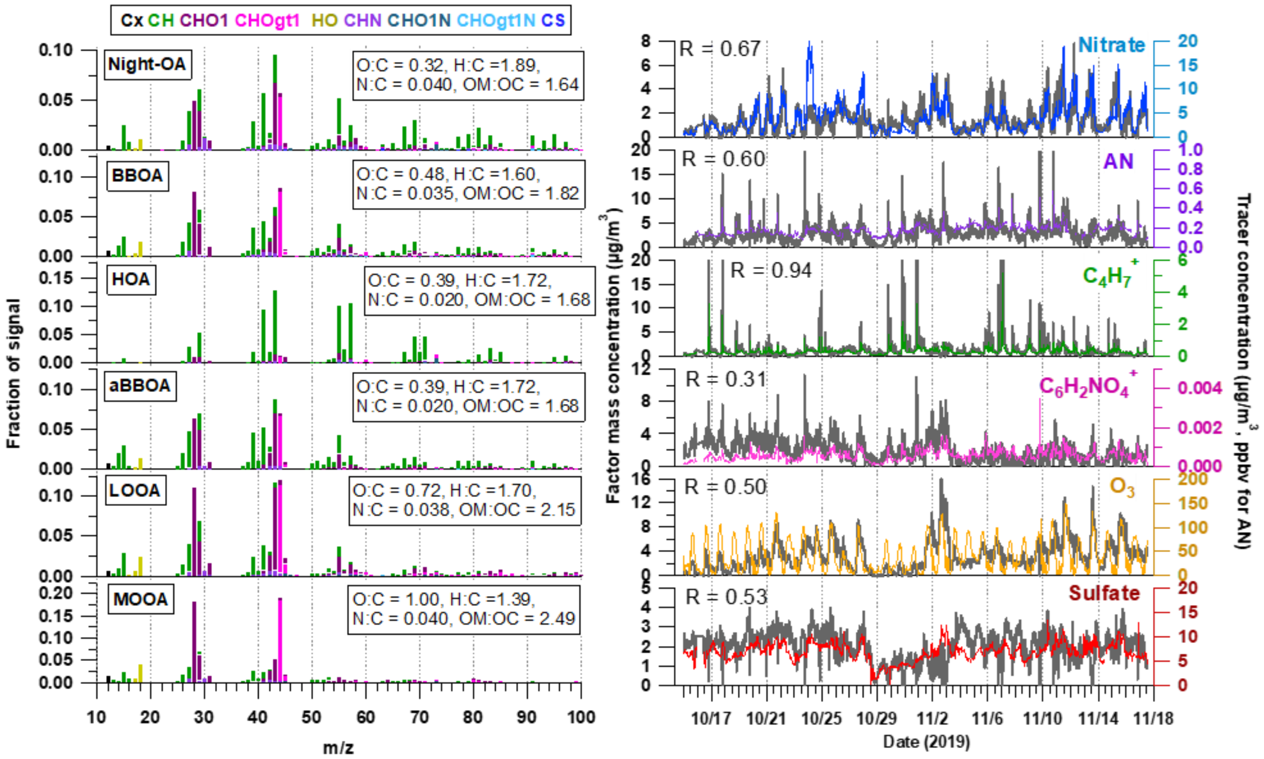

Figure S3. (a) Mass spectral profile in family groups and (b) time series of PMF OA components. Also exhibited are concentration variations of tracer compounds on the right axes.

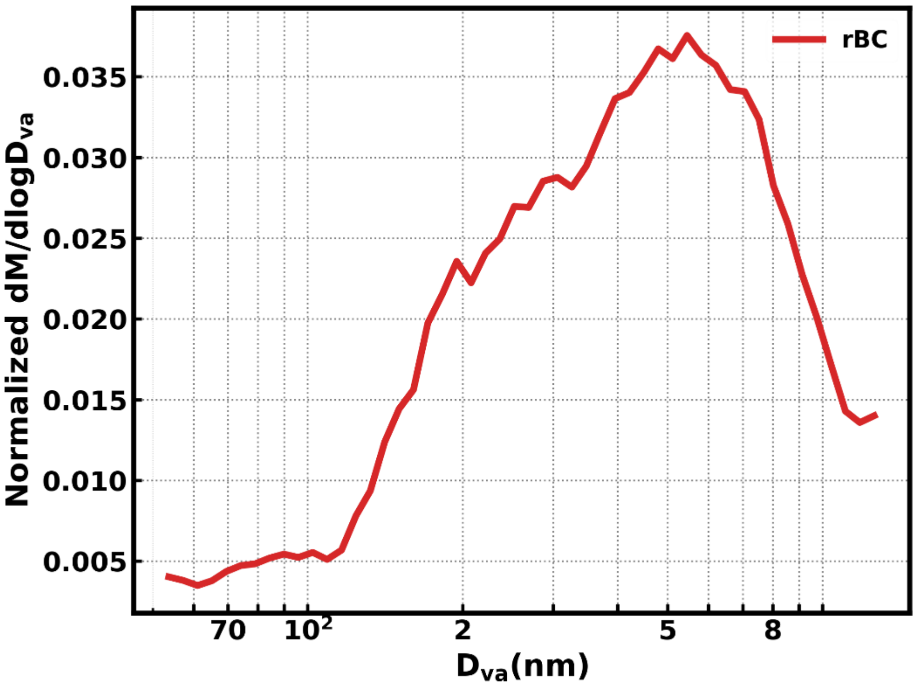

Figure S4. Normalized average Cx distribution measured by the SP-AMS.

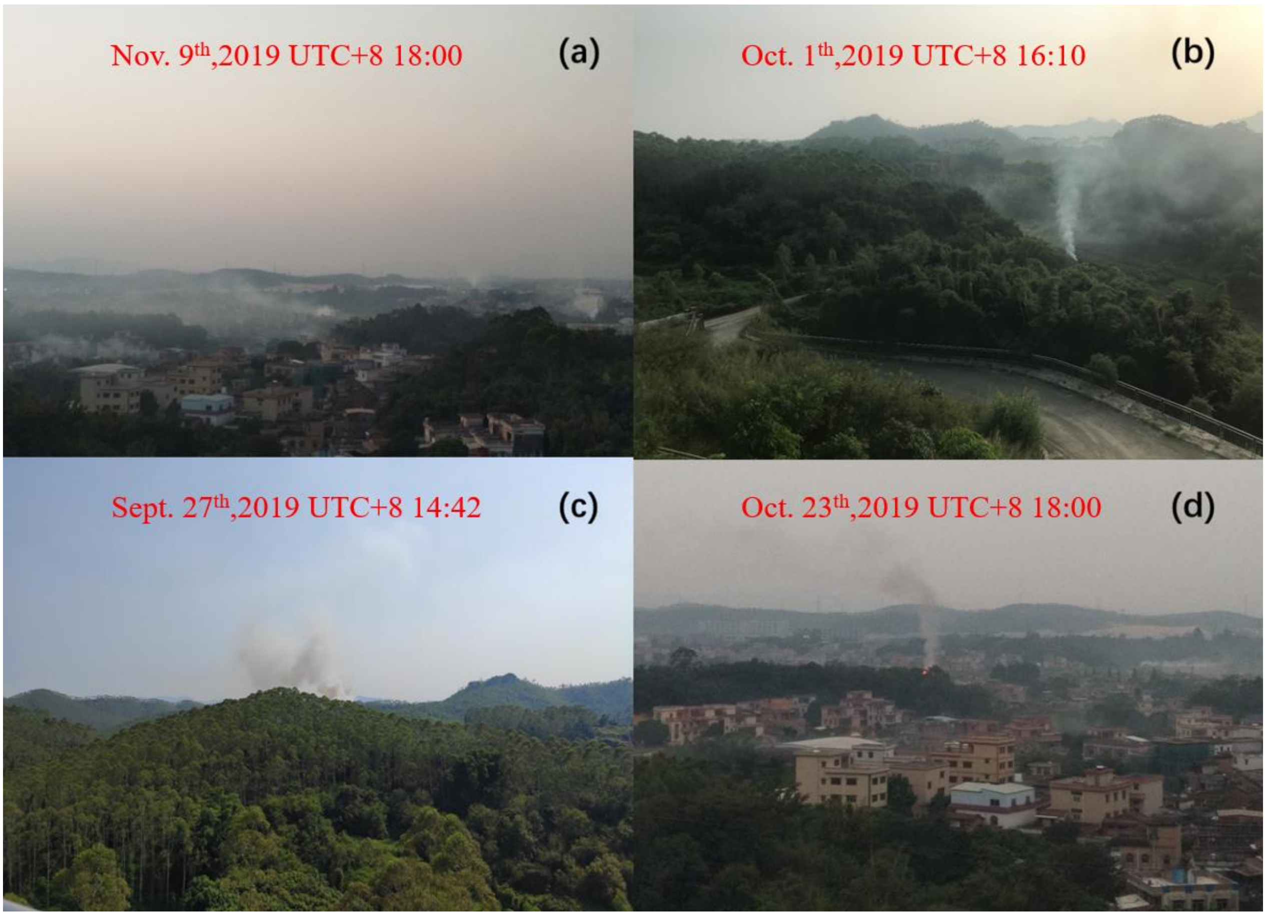

Figure S5. Photoed at at the Heshan supersite during the observation period.

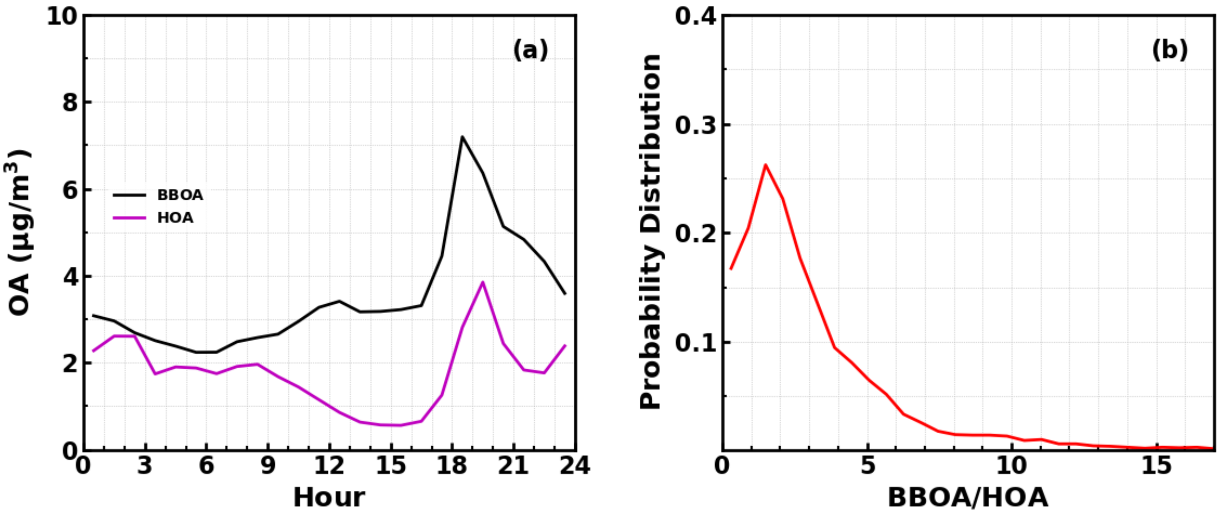

Figure S6. **(a)** Diurnal variations of BBOA and HOA; **(b)** The distribution of BBOA/HOA ratio.

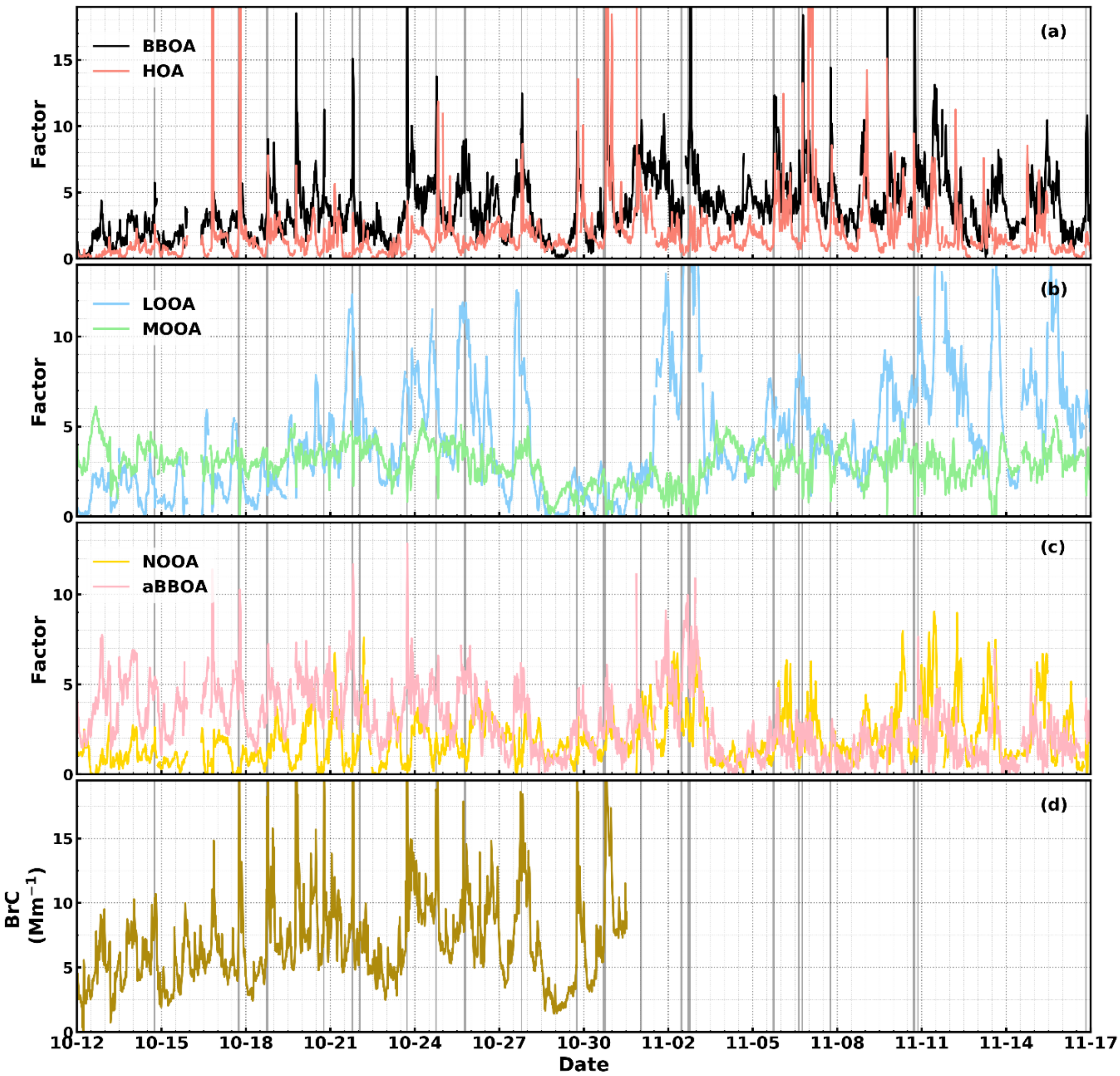

Figure S7. **(a)** to **(c)** Time series of resolved OA factors by SP-AMS measurements, and **(d)** is the derived BrC absorption coefficients at 370 nm. Shaded areas represent identified spikes.

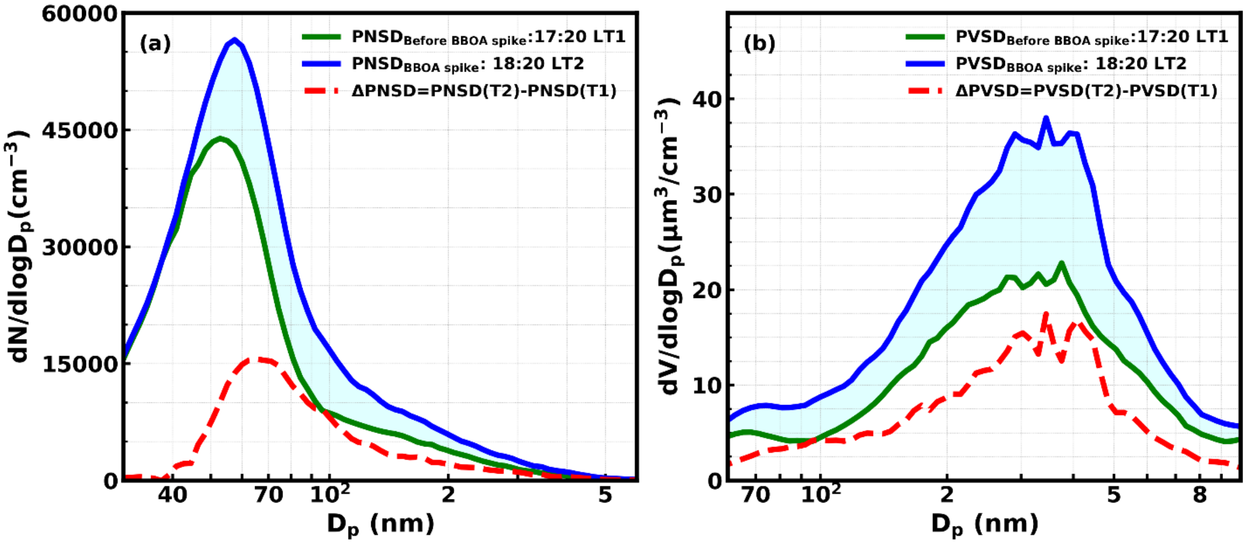

Figure S8. Examples of identifying differences of PNSD (△PNSD) and PVSD (△PVSD)

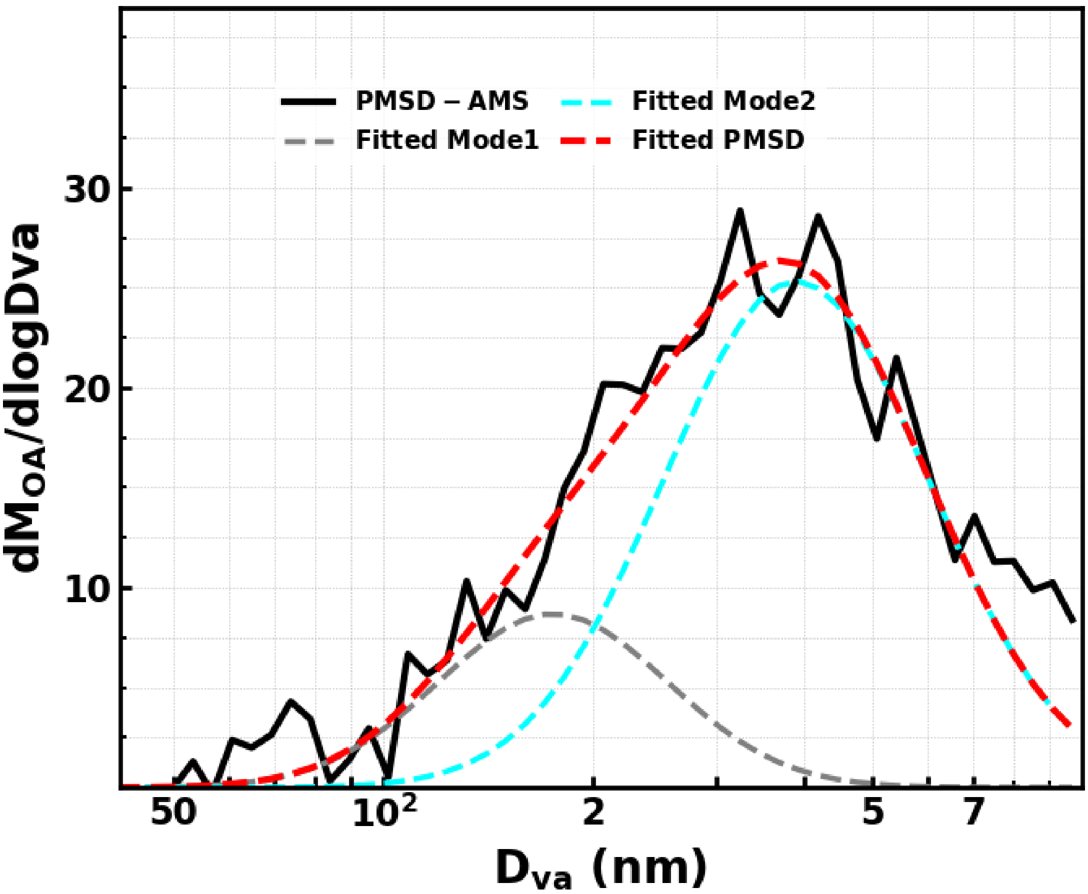

Figure S9. Average OA size distribution differences for spikes in Fig.3b, and the difference can be fitted using two lognormal modes, corresponding to HOA and BBOA respectively.

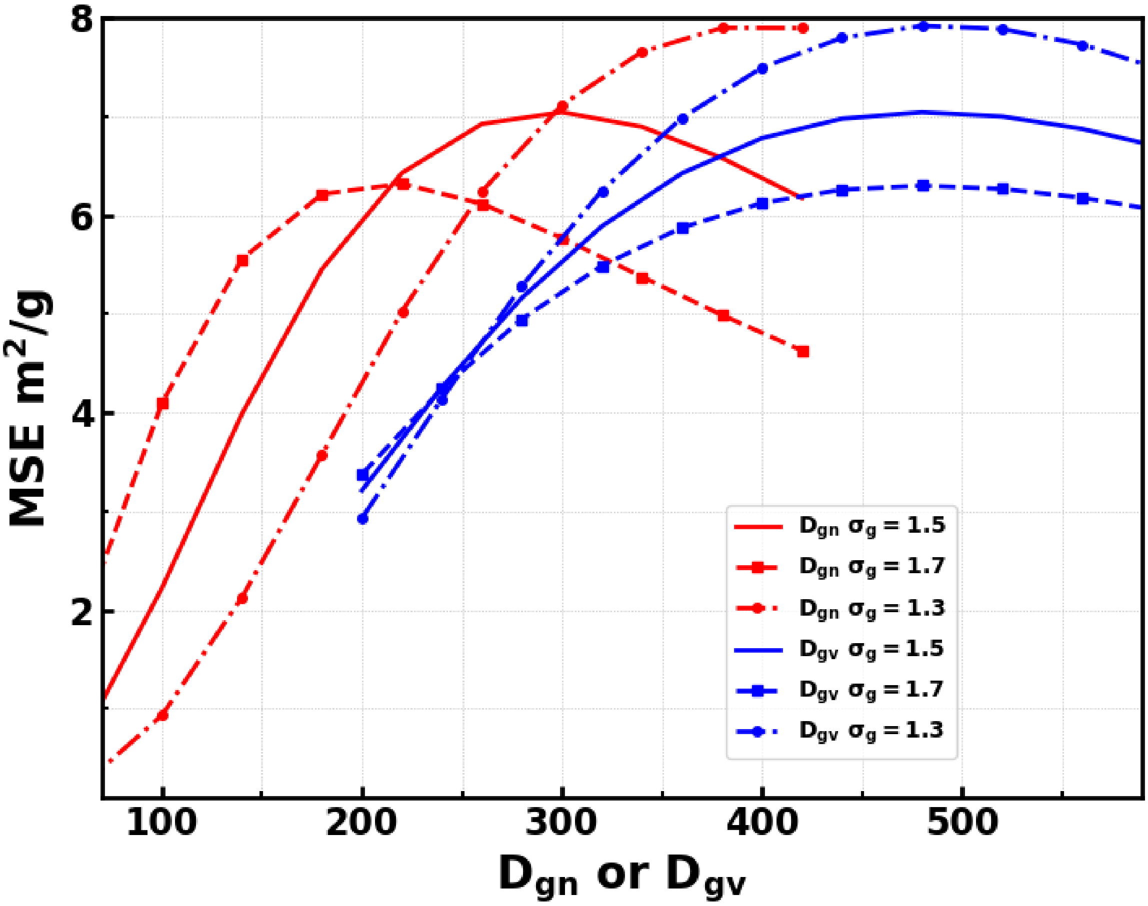

Figure S10. Variations of BBOA MSE under different Dgv, Dgn and $\boldsymbol{\sigma_g}$ conditions.

References

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