1. **Distinct diurnal variation of organic aerosol hygroscopicity and its relationship with oxygenated organic aerosol**

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1. **Aerosol light scattering closure study**
Because that measurements from dry nephelometers are used to estimate $V_{tot}$ for $\kappa_{chem}$ calculations and measured PNSD are used for retrieving $\kappa_{f(RH)}$, the measurement quality of aerosol optical properties and PNSD are important for results in this study. A closure study between measured $\sigma_{sp}$ and that modelled based on measured PNSD with Mie theory (Bohren and Huffman, 2008) is first conducted to double check data quality of used datasets of $\sigma_{sp}$ and PNSD. Measured $\sigma_{sp}$ and $\sigma_{bsp}$ by the nephelometer bears uncertainties associated with angular truncation errors and non-ideal light source (Müller et al., 2011). To achieve consistency between measured and modelled $\sigma_{sp}$, correction factors for measured $\sigma_{sp}$ associated with truncation errors and non-ideal light source are calculated based on parameters for truncation and non-Lambertian illumination correction functions provided by (Müller et al., 2011). For modelling $\sigma_{sp}$ and corresponding correction factors using Mie theory, BC was considered to be half externally and half core-shell mixed with other non-light-absorbing aerosol components. Refractive index and density of BC were assumed to be $1.80 - 0.54i$ and $1.5g cm^{-3}$ (Kuang et al., 2015). Refractive index of non-light-absorbing aerosol components (other than BC) was set to be $1.53 - 10^{-7}i$ (Wex et al., 2002). More details about Mie calculation please refer to Kuang et al. (2015).
The closure results between modelled and measured $\sigma_{sp}$ and $\sigma_{bsp}$ at 525 nm for PM1 and PM10 aerosol particles are shown in Fig. 1. Modelled $\sigma_{bsp}$ for both PM1 and PM10 agree well with the measured $\sigma_{bsp}$, and most points lie between the 20% relative lines. However, Modelled $\sigma_{sp}$ for both PM1 and PM10 are obviously higher than measured $\sigma_{sp}$, and the average relative difference between them for PM10 and PM1 are 22% and 13%, respectively. Considering the measured PNSD by SMPS for particles larger than 200 nm has an uncertainty range of 30% (Wiedensohler et al., 2012), and the measured $\sigma_{sp}$ has an uncertainty of about 9% (Sherman et al., 2015), modelled and measured $\sigma_{sp}$ and $\sigma_{bsp}$ values agree well with each other during this campaign.

2. supplement figures

Figure S1. Comparison between measured and modelled $\sigma_{sp}$ and $\sigma_{bsp}$ at 525 nm, solid red line is the 1:1 line, and red dashed lines are 20% relative lines.
Figure S2. Comparisons between $V_{\text{tot,PNSD}}$ and $V_{\text{tot,Chem}}$ (a), $V_{\text{tot,PNSD}}$ and $V_{\text{tot,Neph}}$ (b), the unit of $V_{\text{tot}}$ is $\mu m^3/cm^3$.

Figure S3. The size-resolved $\sigma_{sp}$ contributions simulated based on the average PNSD of PM10 of period 2.
Figure S4. Examples of PNSD of PM10 and PM1 during fog events and non-fog events.

Figure S5. Size-resolved $\kappa$ distributions which are derived from measured size-segregated chemical compositions during HaChi campaign, colors represent corresponding values of average $\sigma_{sp}$ at 550 nm ($Mm^{-1}$), black solid line is the average size-resolved $\kappa$ distribution and error bars are standard deviations. (reprint from (Kuang et al., 2018))
Figure S6. Normalized size-resolved volume or $\sigma_{sp}$ distribution of PM$_1$ for average PNSDs corresponding to five ranges of aerosol Ångström exponent (0.9-1.1, 1.1-1.3, 1.3-1.5, 1.5-1.7, 1.7-1.9) during this field campaign.
Figure S7. Normalized size-resolved volume or \( \sigma_{sp} \) distribution of PM\(_{1}\) for average PNSDs corresponding to five ranges of aerosol Ångström exponent (0.9-1.1, 1.1-1.3, 1.3-1.5, 1.5-1.7, 1.7-1.9).

Figure S8. x-axis represents mass fraction of nitrate in NR-PM1, and y axis represents the difference between calculated and measured \( k_{chem} \) in Period1.


