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 $\boldsymbol{\kappa_{chem}}$ calculations and measured PNSD are used for retrieving $\kappa_{f(\mathrm{RH})}$, the measurement quality of aerosol optical properties and PNSD are important for results in this study. A closure study between measured σ_{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 σ_{sp} and PNSD. Measured σ_{sp} and σ_{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 σ_{sp}, correction factors for measured σ_{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 σ_{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 $1.80 - 0.54\mathrm{i}$ and $1.5\mathrm{g}\ cm^{-3}$ (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).

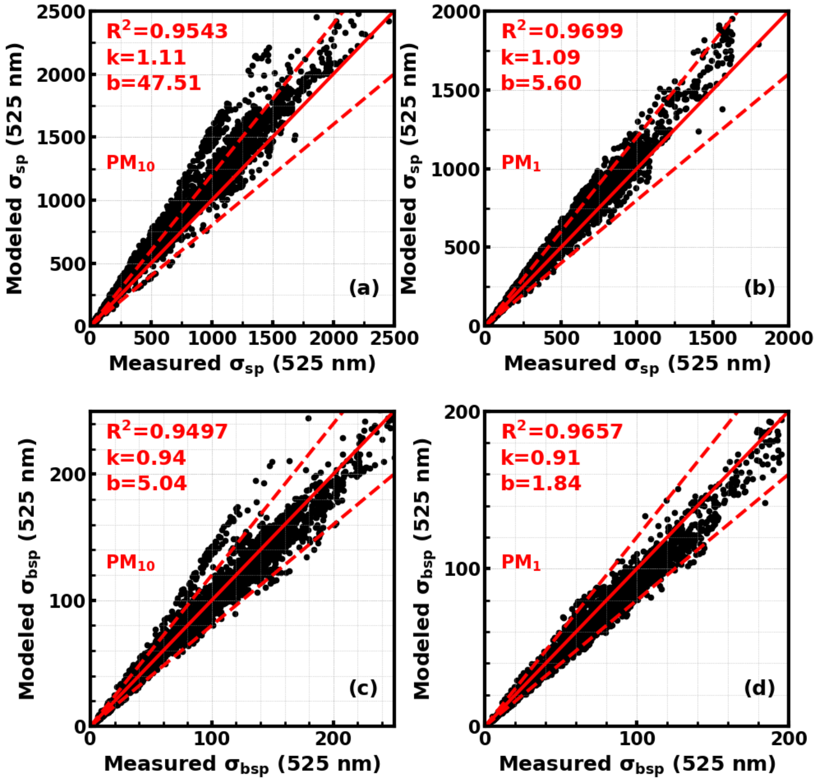

Figure S1. comparison between measured and modelled σ_{sp} and σ_{bsp} at 525 nm, solid read line is the 1:1 line, and red dashed lines are 20% relative lines.

The closure results between modelled and measured σ_{sp} and σ_{bsp} at 525 nm for PM1 and PM10 aerosol particles are shown in Fig.1. Modelled σ_{bsp} for both PM1 and PM10 agree well with the measured σ_{bsp}, and most points line between the 20% relative lines. However, Modelled σ_{sp} for both PM1 and PM10 are obviously higher than measured σ_{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 σ_{sp} has an uncertainty of about 9% (Sherman et al., 2015), modelled and measured σ_{sp} and σ_{bsp} values agree well with each other during this campaign.

2. supplement figures

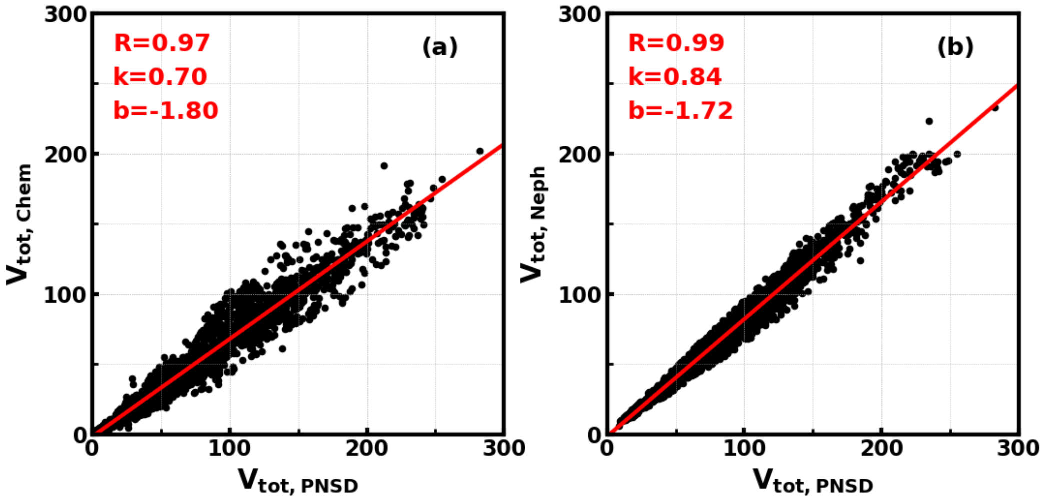

Figure S2. Comparisons between $V_{tot,PNSD}$ and $V_{tot,Chem}$ (a), $V_{tot,PNSD}$ and $V_{tot,Neph}$ (b), the unit of V_{tot} is $\mu m^3/cm^3$.

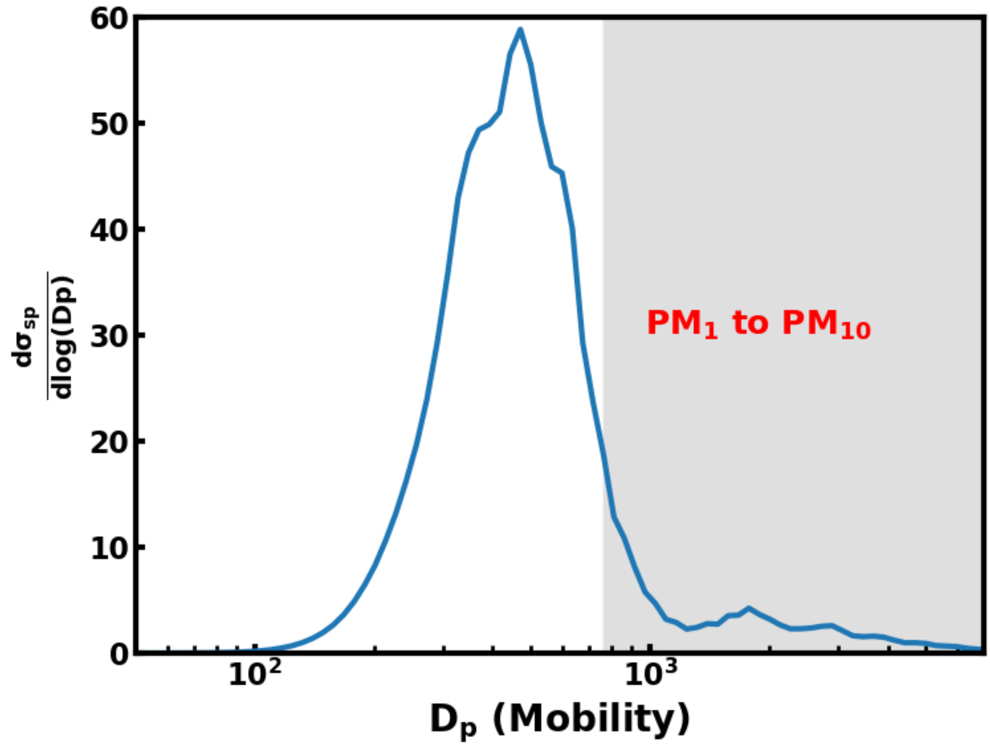

Figure S3. The size-resolved σ_{sp} contributions simulated based on the average PNSD of PM10 of period 2.

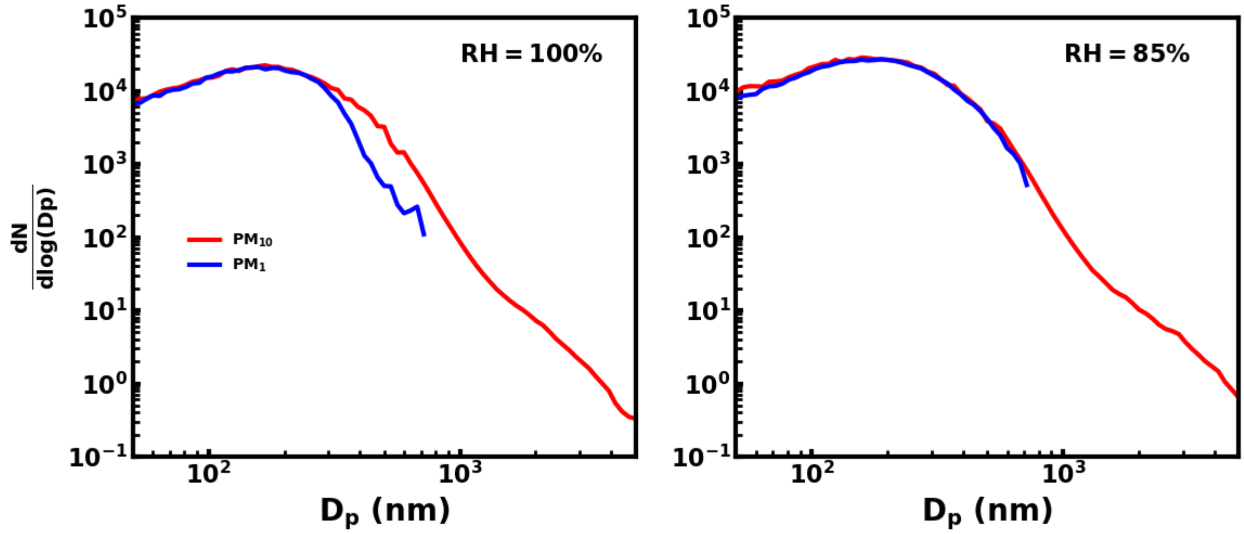

Figure S4. Examples of PNSD of PM10 and PM1 during fog events and non-fog events.

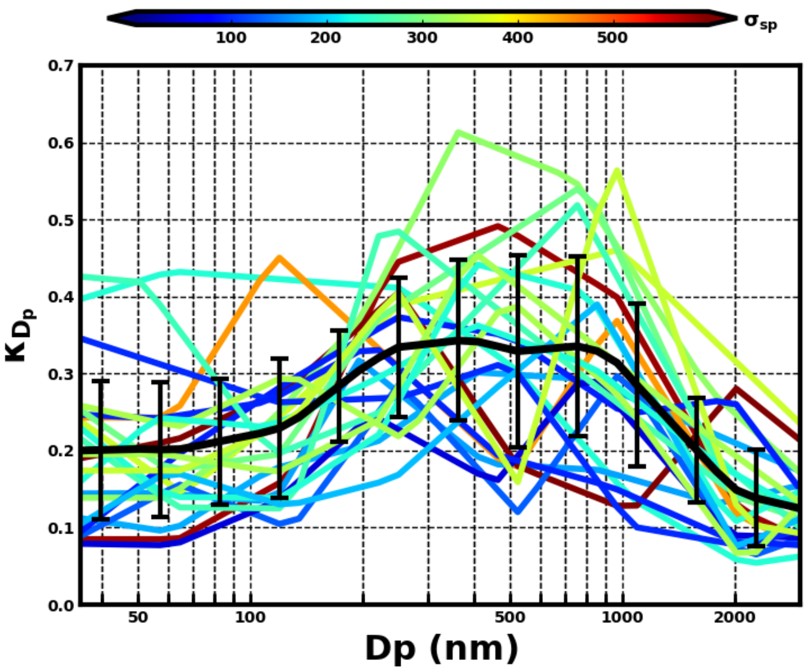

Figure S5. Size-resolved κ distributions which are derived from measured size-segregated chemical compositions during HaChi campaign, colors represent corresponding values of average σ_{sp} at 550 nm (Mm^{-1}), black solid line is the average size-resolved κ distribution and error bars are standard deviations. (reprint from (Kuang et al., 2018))

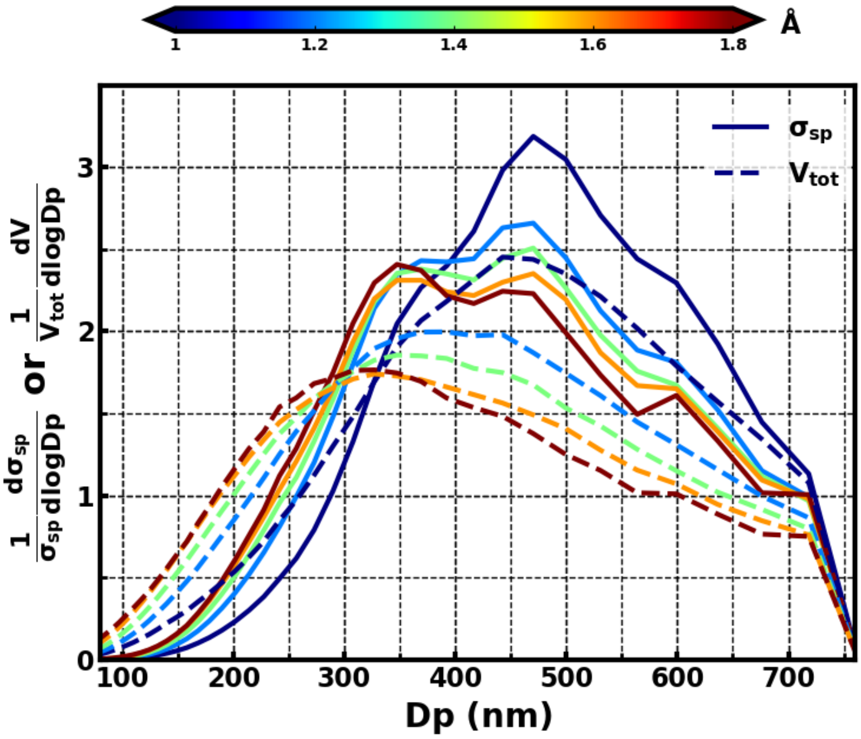

Figure S6. Normalized size-resolved volume or σ_{sp} distribution of PM_1 for average PNSDs corresponding to five ranges of aerosol Ångström exponent (0.9-1.1,1.1-1.3,1.3-1.5,1.5-1.7,1.7-1.9) during this field campaign

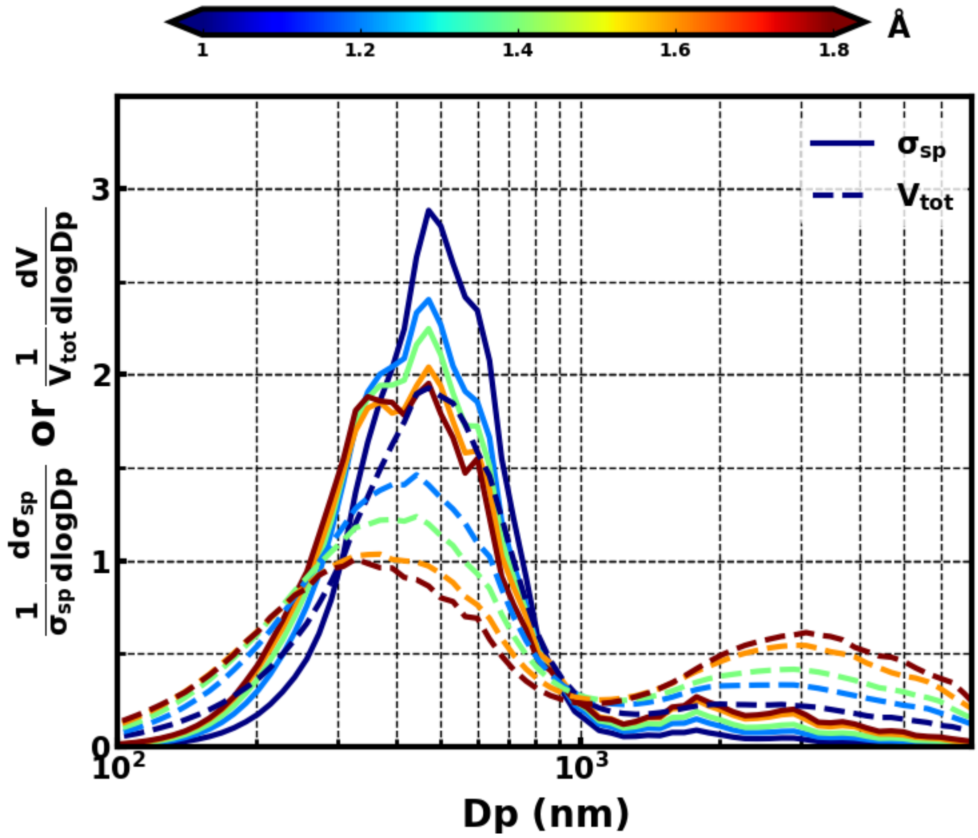

Figure S7. Normalized size-resolved volume or σ_{sp} distribution of PM$_1$ for average PNSDs corresponding to five ranges of aerosol Ångström exponent (0.9-1.1,1.1-1.3,1.3-1.5,1.5-1.7,1.7-1.9)

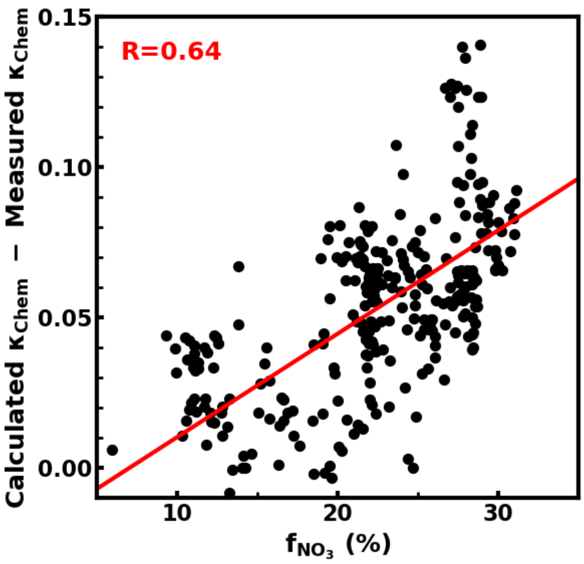

Figure S8. x-axis represents mass fraction of nitrate in NR-PM1, and y axis represents the difference between calculated and measured κ_{chem} in Period1.

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