Supplement for

"Theoretical Framework for Measuring Cloud Effective Supersaturation Fluctuations with an Advanced Optical System"

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1. The reason we choose PM_1 of total aerosol populations as the reference

In clouds, part aerosol activates into cloud droplets, and the rest of them remain as interstitial aerosols. Therefore, the scatterings of total aerosol populations in dry state $\sigma_{sp,all}(\lambda)$ can be expressed:

$$\sigma_{sp,all}(\lambda) = \sigma_{sp,inter}(\lambda) + \sigma_{sp,act}(\lambda) \quad (1)$$

Where $\sigma_{sp,inter}(\lambda)$ and $\sigma_{sp,inter}(\lambda)$ represent scatterings of interstitial aerosols and activated aerosols in dry sate, and λ is the optical wavelength.

If using 1 μm as the threshold of activated aerosols, $\sigma_{sp,inter}(\lambda)$ can be expressed as:

$$\sigma_{sp,inter}(\lambda) = \sigma_{sp,inter,PM_1}(\lambda) \quad (2)$$

Where $\sigma_{sp,inter,PM_1}(\lambda)$ represents scatterings of interstitial aerosols that are PM_1 in dry state, and $\sigma_{sp,inter,PM_{1-2.5}}(\lambda)$ represents scatterings interstitial aerosols that are in the aerodynamic diameter range of 1-2.5 μm. And the $\sigma_{sp,act}(\lambda)$ can be expressed as:

$$\sigma_{sp,act}(\lambda) = \sigma_{sp,act,PM_1}(\lambda) + \sigma_{sp,act,PM_{1-2.5}}(\lambda) + \sigma_{sp,act,PM_{>2.5}}(\lambda) \quad (3)$$

Where $\sigma_{sp,act,PM_1}(\lambda)$ represents scatterings of activated aerosols that are PM_1 in dry state, $\sigma_{sp,act,PM_{1-2.5}}(\lambda)$ represents scatterings of activated aerosols that are in the aerodynamic diameter range of 1-2.5 μm and $\sigma_{sp,act,PM_{>2.5}}(\lambda)$ represents scatterings of activated aerosols that are in the aerodynamic diameter range of >2.5 μm. Therefore, $\sigma_{sp,all}(\lambda)$ can be expressed as:

$$\sigma_{sp,all}(\lambda) = \sigma_{sp,inter,PM_1}(\lambda) + \sigma_{sp,act,PM_1}(\lambda) + \sigma_{sp,act,PM_{1-2.5}}(\lambda) + \sigma_{sp,act,PM_{>2.5}}(\lambda) \quad (4)$$

If a PM_1 impactor were not used after water vapor evaporated for the TSP inlet measurements. Then the ratio $f_{sp} = \sigma_{sp,inter}(\lambda)/\sigma_{sp,all}(\lambda)$ lower than 1 could corresponding to two scenarios: (1) part of submicron aerosols have activated with $\sigma_{sp,act,PM_{>1}}(\lambda)$ are negligible; (2) no aerosols are activated with $\sigma_{sp,act,PM_{>1}}(\lambda)$ are not negligible. That means, we could observe f_{sp} lower than 1 under both subsaturated conditions and supersaturated conditions, and this would obscure the D_a retrievals in cloud conditions, especially at lower supersaturations, when D_a is higher and $\sigma_{sp,inter}(\lambda)$ itself is relatively small. However, if a PM1 impactor is placed downstream of the inlet and upstream of the two nephelometers (or other optical instruments), the observed f_{sp} can be expressed:

$$f_{sp} = \sigma_{sp,inter,PM_1}(\lambda)/(\sigma_{sp,inter,PM_1}(\lambda) + \sigma_{sp,act,PM_1}(\lambda)) \quad (5)$$

f_{sp} be lower than 1 could only be caused by activation of submicron aerosols, therefore, facilitate the accurate retrieval of D_a.

If using 2.5 μm as the threshold of activated aerosols, $\sigma_{sp,inter}(\lambda)$ can be expressed as:

$$\sigma_{sp,inter}(\lambda) = \sigma_{sp,inter,PM_1}(\lambda) + \sigma_{sp,inter,PM_{1-2.5}}(\lambda) \quad (6)$$

the $\sigma_{sp,act}(\lambda)$ can be expressed as:

$$\sigma_{sp,act}(\lambda) = \sigma_{sp,act,PM_1}(\lambda) + \sigma_{sp,act,PM_{1-2.5}}(\lambda) + \sigma_{sp,act,PM_{>2.5}}(\lambda) \quad (7)$$

A $PM_{2.5}$ impactor downstream of TSP inlet after water evaporates would eliminate the influences of $\sigma_{sp,act,PM_{>2.5}}(\lambda)$. However, as demonstrated in Kuang et al. (2018), the $\sigma_{sp,all}(\lambda)$ are not sensitive to changes in super-micron aerosols therefore it would be better if only submicron aerosols are included in observing the scattering fractions of interstitial aerosols and benefits for accurate retrieval of D_a. Therefore, no matter using 1 or 2.5 μm as the threshold, the PM_1 impactor are suggested downstream of the inlet system after heating.

As mentioned, In the concept design of Sect.4 of the manuscript, the interior PM_1 impactor was placed downstream of the inlet system where RH of sample air was heated down to 70%. RH down to 70% is to make sure selected aerosols using the PM1 impactor are very close to aerosols populations of PM_1 in dry state based on the investigates of impacts of aerosol hygroscopic growth on cut-off size shift of impactors (Xu et al., 2024).

2. Simulation details of the relationships between D_a and f_{sp}

The particle number size distributions (PNSDs) in dry state, which range from about 10 nm to 10 μm, were jointly measured by a Twin Differential Mobility Particle Sizer (TDMPS, Leibniz-Institute for Tropospheric Research, Germany; Birmili et al. (1999)) or a scanning mobility particle size spectrometer (SMPS) and an Aerodynamic Particle Sizer (APS, TSI Inc., Model 3321) in six field campaigns conducted on the North China Plain which are detailed in Kuang et al. (2018). The mass concentrations of black carbon (BC) were measured using a Multi-Angle Absorption Photometer (MAAP Model 5012, Thermo, Inc., Waltham, MA USA) or an aethalometer (AE33) (Drinovec et al., 2015) in these field campaigns. For each paired PNSD and BC mass concentration, the size distribution of PM_1 (and the penetration curve shape from Gussman et al. (2002) was also included for considering the non-ideality cutoff of the impactor, and assuming aerosol density of 1.6 g/cm^3 for converting aerodynamic diameter to mobility diameter) :

$$\mathrm{PNSD}\ (\mathrm{D_p})_{PM_1} = \mathrm{PNSD}\ (\mathrm{D_p}) \times \mathrm{R}(\mathrm{D_p}) \quad (8)$$

Where R(Dp) is the penetration ratio of aerosols as a function of particle diameter D_p of the PM_1 impactor.

One hundred size-resolved activation curves are produced for each PNSD $(Dp)_{PM_1}$ using the following formula:

$$AR(D_p)=\frac{MAF}{2}\left(1+\text{erf}\left(\frac{D_p\text{-}D_a}{\sqrt{2\pi}\sigma}\right)\right) \quad (9)$$

Where $AR(D_p)$ was the size-resolved activation ratios, MAF is the maximum activation fraction and D_a is critical activation diameter, σ is associated with the slope of the curve near D_a. This formula was previously proposed by (Rose et al., 2008) to fit the AR measurements and widely used in AR parameterizations (Tao et al., 2018). The random ranges of MAF, D_a and σ are 0.6-1, 80-800 nm, and 10-100 to produced enough types of activation curves for each PNSD $(Dp)_{PM_1}$.

Therefore, the PNSD of interstitial aerosols can be calculated as the following:

$$\text{PNSD}\ (D_p)_{PM_1,inter} = \text{PNSD}\ (D_p)_{PM_1} \times (1 - AR(D_p))$$

Therefore, the $\sigma_{sp,PM_1,all}(dry,\lambda)$ and $\sigma_{sp,PM_1,inter}(dry,\ \lambda)$ at wavelengths of 450 nm, 525 nm and 636 nm for all PNSD $(D_p)_{PM_1,inter}$ and PNSD $(D_p)_{PM_1}$ can be calculated using the Mie theory calculations with the BC mass are distributed based on $AR(D_p)$ and the shape of black carbon mass size distributions are consistent with the one used in simulations of Kuang et al. (2017) assuming fractions of BC mass that are externally mixed is 0.5. Details about the Mie theory calculations can also be found in Kuang et al. (2017). With these configurations, more than million pairs of f_{sp} and D_a are simulated.

3. Other Supplementary Figures

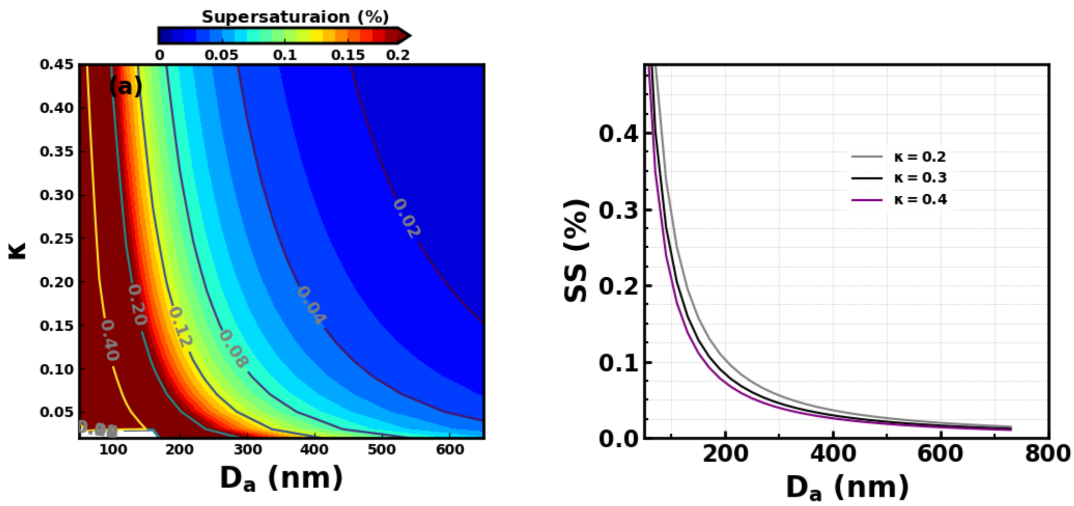

Figure S1. **(a)** Supersaturation (SS) variations under different D_a and κ scenarios; **(b)** The variations SS as a function of D_a for constant κ values of 0.2,0.3,0.4.

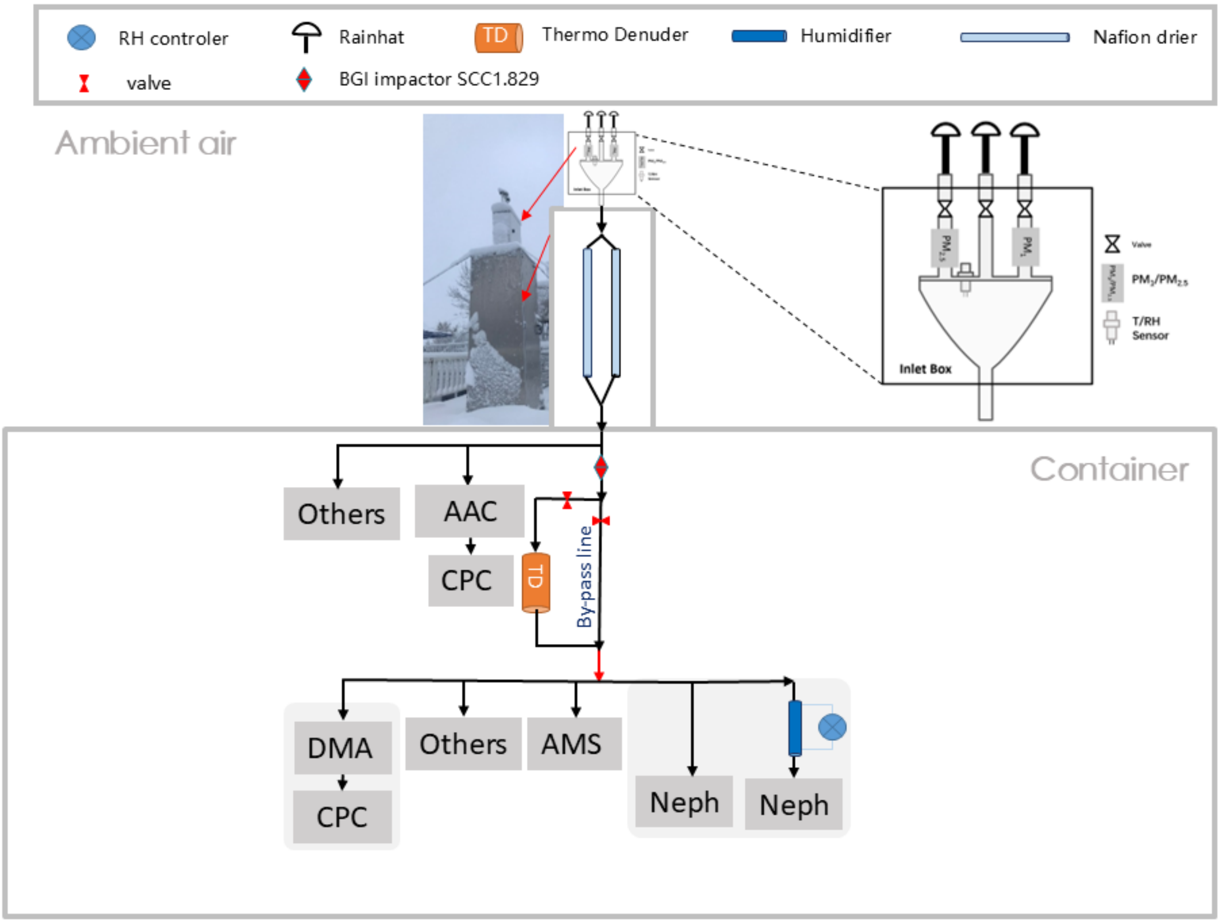

Figure S2. Schematic of instrument setup during the AQ-SOFAR campaign, with aerosol size distributions are measured using Aerodynamic Aerosol Classifier (AAC) and Differential Mobility Analyzer (DMA) coupled with Condensation Particle Counters (CPC, TSI 3076 and 3075), and Neph represents nephelometer.

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