A Method to Obtain Scattering Phase Function based on Particle Size Distribution and Refractive Index Retrieved from Aurora 4000 Multi-angle Scattering Measurements: A Numerical Study

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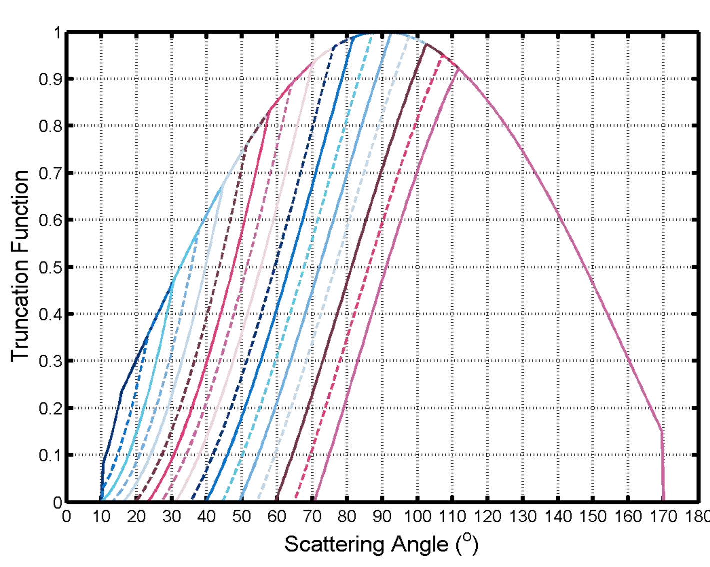
Introduction

Scattering phase function of aerosol particles is crucial to the accurate estimation of aerosol direct radiative forcing. One feasible and promising way to obtain scattering phase function on a long-term scale with satisfying temporal resolution is based upon the particle size distribution and the refractive index simultaneously retrieved from multi-wavelength multi-angle scattering detected by Aurora 4000, a commercially available and easily operated, as well as lab- and field-deployable instrument. In this study, a retrieval algorithm was specifically developed for Aurora 4000 measurements, using the regularization method. The algorithm was systematically evaluated for its applicability, capability and limitation by a series of numerical experiments, in order to be prepared for its application in next field studies.

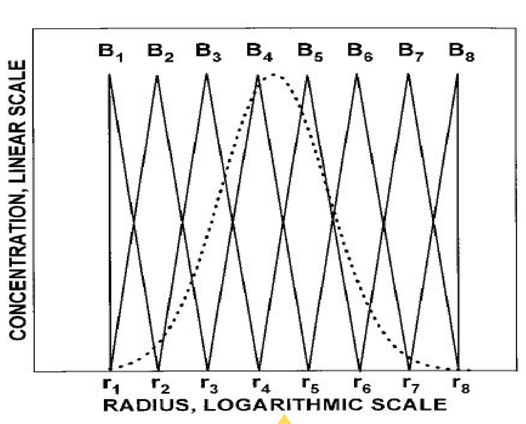
Methodology

1. Instrument

The Aurora 4000 polar nephelometer (Acoem Ecotech, Australia) is capable of measuring light scattered by aerosol particles at multi-angle ranges (Teri et al., 2022) at 3 wavelengths (450, 525 and 635 nm), but subject to an illumination function $Z(\theta,\alpha)$ deviated from the ideal condition (Müller et al., 2012).



2. Retrieval Algorithm



B-spline base functions (Müller et al., 1999)

Mie theory (Bohren and Huffman, 1998)

$$\boldsymbol{\sigma} = \mathbf{A}\mathbf{w} + \boldsymbol{\varepsilon}$$

Regularization method Müller et al. (1999)

Scattering coefficients $\sigma(\alpha,\lambda)$ at 3 wavelengths and 17 angle ranges (α~180°, α starting from 10° to 90°)

$$\mathbf{w} = (\mathbf{A}^{\mathrm{T}}\mathbf{A} + \gamma\mathbf{H})^{-1}\mathbf{A}^{\mathrm{T}}\boldsymbol{\sigma}$$

For a given m, the selection of γ is performed with the widely used generalized cross-validation (GCV) method. With selected γ, a set of **w** (thereby the PSDs) can be obtained for a given inversion window.

(1) Inversion windows: in total 126
 Lower limits: [0.01, 0.014, 0.02, 0.03, 0.05, 0.07, 0.1, 0.14, 0.2] μm
 Upper limits: [0.35, 0.5, 0.7, 1, 1.4, 2, 3, 5, 7, 10, 14, 20, 30, 50] μm

(2) Developed filtering method
 a. Perform a four-step procedure of physical constraints for each solution
 b. Perform a two-step procedure of optical constraints for each solution
 c. Treat all solutions from all inversion windows together
 d. Average the remaining solutions to separately obtain the three PSDs

(3) Merging method
 Merge the results from directly inverted PNSD, PSSD and PVSD to give the final solution, using a sigmoidal function as weighting factors

(4) Estimating n
 a. Cauchy equation (Jenkins and White, 2001): $n = n_{ref} + B(\frac{1}{\lambda^2} - \frac{1}{\lambda_{ref}^2})$
 n_{ref}: 1.33~1.93 at 525 nm with an interval of 0.01
 B: -9000~24000 nm^2 with an interval of 3000 nm^2
 b. Obtain the PSD for each m=n+ik following (1)-(3)
 k is assumed 0 at all wavelengths
 c. Filter solutions and average them to obtain the final PSD and n

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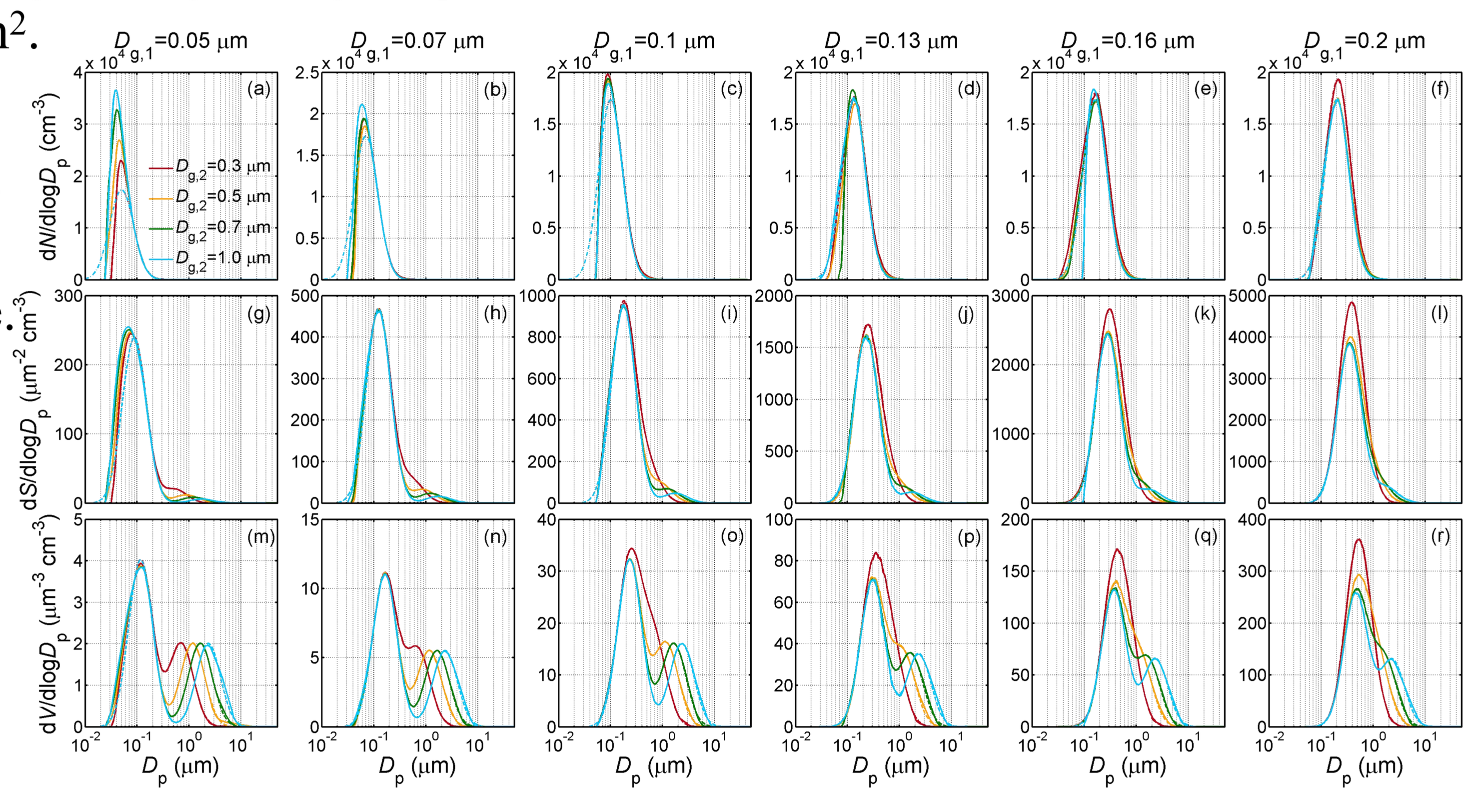
Results & Discussion

1. Retrievals for purely scattering aerosol particles

The PSDs can be reliably retrieved, with either known n or unknown n, for:
a. monomodal ($12D_g \times 4\sigma_g$) log-normal distributions with $7n_0 \times 12B_0$,
b. bimodal ($6D_{g,1} \times 4D_{g,2} \times 1\sigma_g \times 3r_{peak}$) log-normal distributions with n_0=1.53, B_0=6000 nm^2.

Examples of bimodal cases with known n are given here. Solid and dashed lines represent inverted and true PSDs.

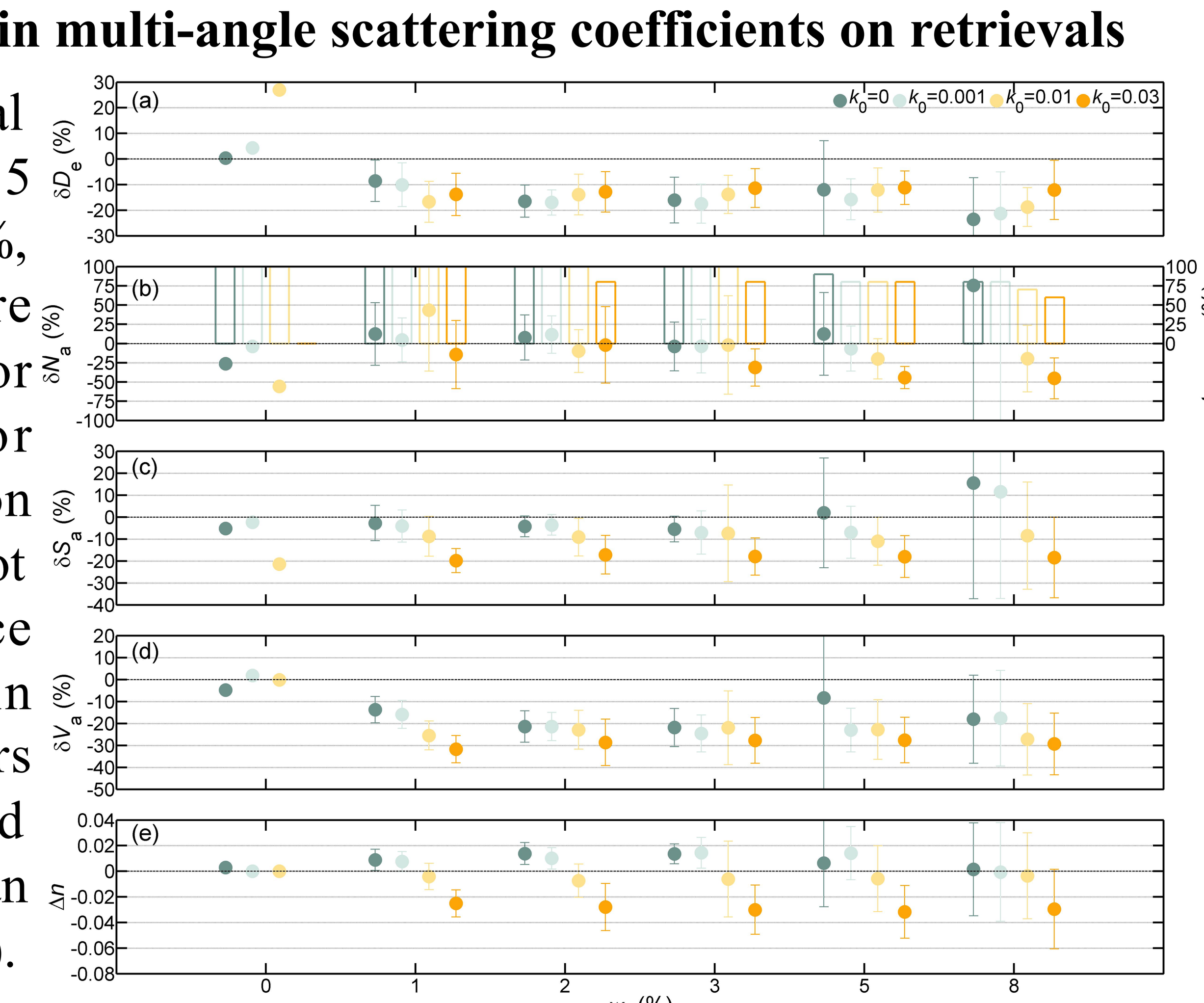


2. Retrievals for partially absorbing aerosol particles

With known m, the PSDs can be well retrieved even for strongly absorbing aerosol particles (k=0.5). With only known n or unknown n for an estimation, retrievals of the PSDs and n are limited to slightly absorbing aerosol particles (k≤0.01).

3. The effect of errors in multi-angle scattering coefficients on retrievals

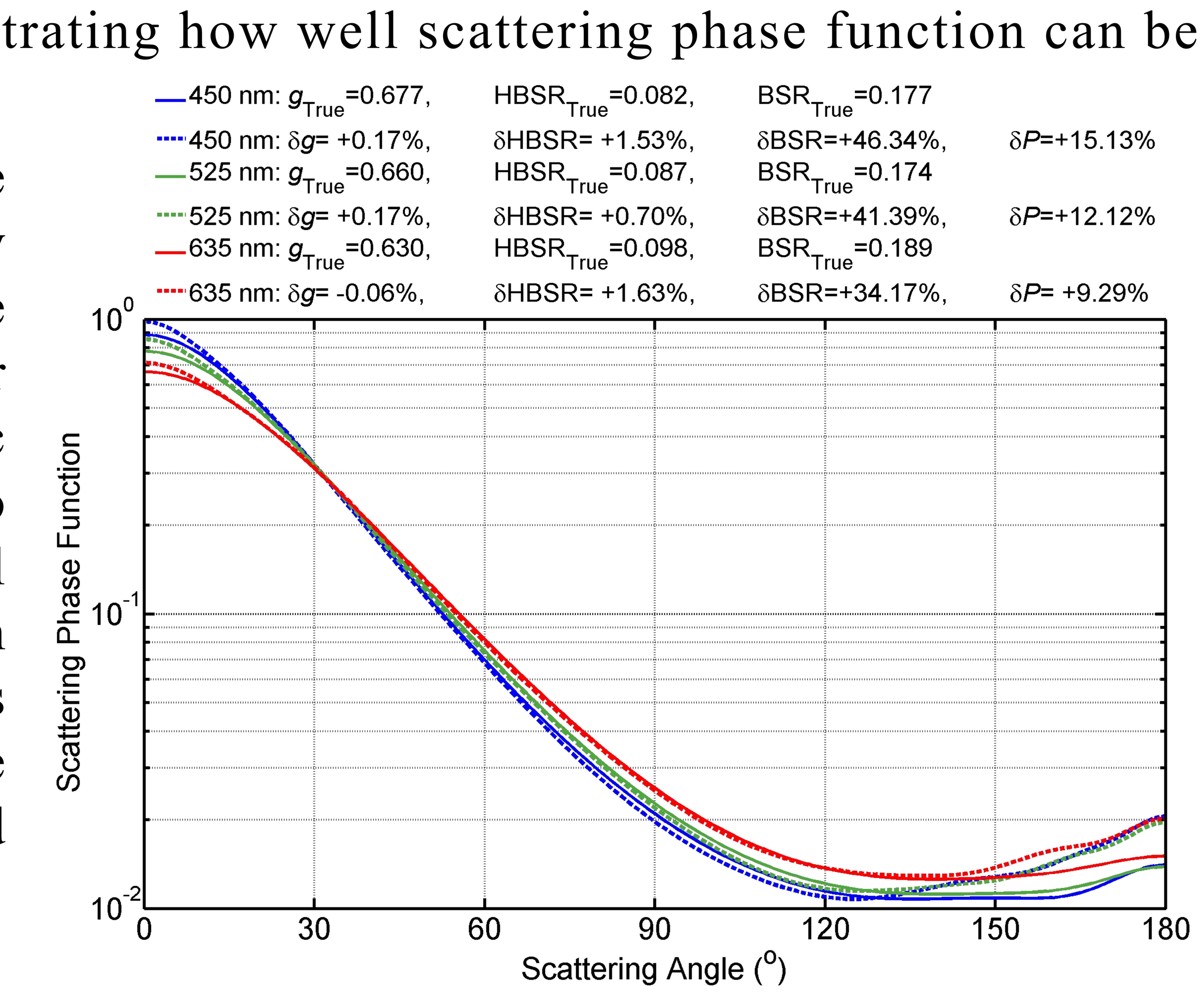
Random errors in optical measurements within 5 ranges ($\pm\psi_s$=±1%, ±2%, ±3%, ±5%, ±8%) were tested, with 10 runs for each error range. For k≤0.01, the introduction of optical errors does not undermine performance of the algorithm within ±3%, reported as errors for Aurora data averaged over a longer period than 1-min (Teri et al., 2022).



4. The assessment of aerosol scattering phase function

Relative errors in backscattering ratio (BSR) and the defined parameter to describe relative errors in scattering phase function (δP) are found to be good indicators for demonstrating how well scattering phase function can be captured.

In contrast, relative errors in the widely used parameters of the asymmetry parameter (g) and hemispheric backscattering ratio (HBSR) remain still quite small even when considerable deviations can be observed in the forward and backward scattering regimes.



5. Summary

- The retrieval algorithm has exhibited a successful performance in deriving the PSDs and n, and consequently scattering phase function
- It is applicable to aerosol particles from purely scattering to slightly absorbing (k≤0.01), both without and with measurement errors (within ±3%)
- No priori assumptions about the shape of the PSDs and n values are needed