Accurate retrieval of pure black carbon aerosol properties including light absorption from polarization-resolved in situ measurements of light scattering

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Probing angular and polarization dependence of light scattering together with polarimetric retrievals play a crucial role in remote sensing of aerosol size distribution and light absorption. In particular, accurate retrieval absorption by black carbon remains a main challenge, as demonstrated by Schuster et al. (2019) in controlled laboratory experiments, partly due to simplified aerosol representation in standard retrieval algorithms. Here we demonstrate that considering non-spherical shape reduces uncertainty and bias of aerosol property retrievals for BC particles.

We used PSL (spherical, non-absorbing), nigrosin (spherical, absorbing) and surrogates of black carbon particles (non-spherical aggregates, absorbing) as test aerosols. An Aerodynamic Aerosol Classifier was used to produce unimodal size distributions. Phase function and polarized phase function were measured using a laser imaging type nephelometer (Moallemi et al., 2023). Aerosol properties were independently measured using a condensation particle counter, scanning mobility particle sizer, aerosol particle mass analyzer, and photo-acoustic absorption photometer.

Optical forward calculations were either done using Mie theory for spheres or using Multi-Sphere T-Matrix (MSTM) calculations for BC surrogates. Property retrievals were done by fitting the measured phase function data using above forward kernel and aerosol properties as fit parameters.

For spherical homogeneous particles (PSL and nigrosin), agreement with independent data was excellent with RMSE of around $\pm 1.95\%$ for modal diameter, $\pm 7.99\%$ for number concentration. Retrieval of complex refractive index was also in good agreement with literature data with RMSE of ± 0.02 for real part and ± 0.04 for imaginary part. A RMSE of $\pm 6.13\%$ achieved for the absorption coefficient also falls within uncertainty of the independent measurement.

While using Mie kernel does not yield a satisfactory match to phase functions measured for nonspherical BC aggregates (blue line in Fig. 1) leading to systematic biases in retrieved aerosol parameters. In contrast, using the MSTM kernel provided a good fit (red lines in Fig. 1) and resulted in unbiased retrieval of aerosol properties including absorption within uncertainty. Future work will focus on coated BC and on ambient samples to assess the potential of reducing retrieval uncertainty for atmospheric aerosol by including a BC particle component in the aerosol model and optical forward kernel.

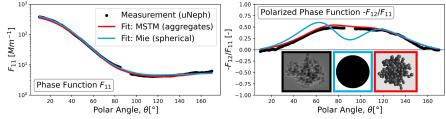


Fig. 1 Measured and fitted phase functions for size-selected pure BC with an aggregate shape. The results demonstrate that a simple aggregate model (monodisperse primary spheres) is sufficient for accurate BC property retrieval given simple BC aerosol samples.

- [1] Moallemi, A., et al. Atmos. Meas. Tech., **2023**, 16, 3653-3678.
- [2] Schuster, G. L., Espinosa, W. R., et al. Remote Sens., 2019, 11, 498