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Comparing aerosol refractive indices retrieved from full distribution and size- and mass-selected measurements

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ABSTRACT

Refractive index retrievals (also termed inverse Mie methods or optical closure) have seen considerable use as a method to extract the refractive index of aerosol particles from measured optical properties. Retrievals of an aerosol refractive index use one of two primary methods: 1) measurements of the extinction, absorption and/or scattering cross-sections or efficiencies of size- (and mass-) selected particles for mass-mobility refractive index retrievals (MM-RIR) or 2) measurements of aerosol size distributions and a combination of the extinction, absorption and/or scattering coefficients for full distribution refractive index retrievals (FD-RIR). These two methods were compared in this study using pure and mixtures of ammonium sulfate (AS) and nigrosin aerosol, which constitute a non-absorbing and absorbing material, respectively. The results indicate that the retrieved complex refractive index values are correlated to the amount of nigrosin in the aerosol but can be highly variable with differences in the real and imaginary components that range between −0.002 and 0.216 and −0.013 and 0.086; the average and standard deviation of the differences are 0.046 ± 0.046 and 0.023 ± 0.033, respectively. Forward calculation of the optical properties yielded average absolute values of the relative deviation of ≈ 15% and ≈ 26% for FD-RIR data using the MM-RIR values and contrariwise. The range of retrieved refractive indices were used to calculate the normalized global average aerosol radiative forcing of a model accumulation mode remote continental aerosol. Deviations using the refractive indices of the pure materials range from 9% to 32% for AS and 27% to 45% for nigrosin. For mixtures of nigrosin and AS, deviations were all > 100% and not always able to capture the correct direction of the forcing; i.e., positive versus negative.

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Abbreviations and Variable Definitions

APM	Aerosol particle mass analyzer
AS	Ammonium sulfate
C_{abs}	Absorption cross-section (m^2)
C_{ext}	Extinction cross-section (m^2)
C_{scat}	Scattering cross-section (m^2)
CPC	Condensation particle counter
CRD	Cavity ring-down spectrometer
CV	Coefficient of variation (%)
D_{fm}	Mass-mobility scaling exponent
D_m	Mobility diameter (nm)
DMA	Differential mobility analyzer
FD-RIR	Full Distribution refractive index retrieval
k_0	Prefactor for mass-mobility scaling relationship
m	Complex refractive index
m_p	Particle mass (g)
m_{eff}	Effective particle mass (g)
MM-RIR	Mass-mobility refractive index retrieval
N	Number density of aerosol particles (m^{-3})
n	Real component of the complex refractive index
k	Imaginary component of the complex refractive index
LOD	Limit of detection
PA	Photoacoustic spectrometer
q	Net charge on an aerosol particle
Q_{abs}	Absorption efficiency
Q_{ext}	Extinction efficiency
Q_{scat}	Scattering efficiency
RH	Relative humidity (%)
SMPS	Scanning mobility particle sizer
w_{AS}	Ammonium sulfate mass fraction
x	Real component of the microphone/power meter response
y	Imaginary component of the microphone/power meter response
α_{abs}	Absorption coefficient (m^{-1})
α_{bscat}	Backscattering coefficient (m^{-1})
α_{ext}	Extinction coefficient (m^{-1})
α_{scat}	Scattering coefficient (m^{-1})

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λ	Wavelength (nm)
ρ	Mass density (g cm^{-3})
ρ_{eff}	Effective mass density (g cm^{-3})
σ	Uncertainty level from either a standard deviation or other statistical method. At 1σ, range contains ≈ 68.2% of observations with cumulative percentiles spanning 15.9% to 84.1%.
σ_p	Mass distribution width
σ_{eff}	Effective mass distribution width
χ^2	Merit function for determination of refractive indices

1. Introduction

Aerosols directly affect the radiation budget of the earth through the absorption and scattering of incoming solar radiation. Accurate quantification of the radiative forcing magnitude requires knowledge of the spatial (latitude, longitude and altitude) and temporal distributions of aerosol particles and their corresponding chemical, physical, and optical properties. Satellite observations provide excellent spatial and temporal resolution but are limited to observations of columnar optical depth which does not provide speciation or vertical profiles. Field measurements from both ground-based stations and airplanes can fill these data gaps with detailed measurements of the physical properties of aerosols (e.g., optical, morphological, chemical, etc.) but are limited in temporal and spatial resolution. Models, such as the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) [1], the optical properties of aerosols and clouds (OPAC) software package [2] or the Generalized Retrieval of Aerosol and Surface Properties (GRASP) [3] serve as the bridge between satellite and in situ measurements, but also aid in the prediction of future radiative forcing scenarios. These models calculate aerosol optical properties using Mie theory with individual aerosol components (e.g., sulfate, organic carbon, black carbon, mineral dust, etc.) parameterized by their refractive indices, hygroscopicity and typical size distributions.

Because models rely heavily on refractive indices to calculate aerosol optical properties, many investigations have focused on the "inverse problem" [4] of empirically retrieving the complex refractive index (m)

$$m = n + ik \tag{1}$$

from measured optical and morphological data by either: 1) size- (and mass-) selecting particles and measuring some combination of the extinction, scattering and absorption efficiencies (Q_{ext}, Q_{scat} and Q_{abs}, respectively) or cross-sections (C_{ext}, C_{scat} and C_{abs}, respectively) [5–24] – efficiencies are the ratio of the optical cross-section to physical cross-section – or 2) using the full distribution of aerosol particles and measuring the size distribution and at least two of the extinction (α_{ext}), scattering (α_{scat}), backscattering (α_{bscat}) and/or absorption (α_{abs}) coefficients [25–42]. Chemical species data have also been used to calculate an effective refractive index that is then compared to measured optical data [35,43]. For the remainder of this manuscript, the terms retrieved and calculated (and their grammatically correct derivatives) are used to refer to an inverse method where refractive indices are "retrieved" from measured optical values and a forward method where optical values are "calculated" from refractive indices. Following this framework, the two methods mentioned above will be referred to as: 1) mass-mobility refractive index retrievals (MM-RIR) and 2) full distribution refractive index retrievals (FD-RIR), respectively. While these retrievals are typically performed at a single wavelength, multi-wavelength retrievals of extinction spectra have been performed utilizing the Kramers-Kronig dispersion relationship as an additional constraint [44–53].

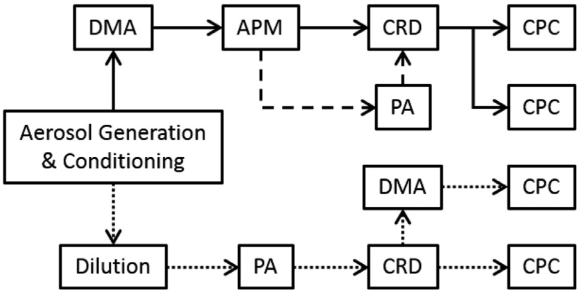

Fig. 1. Block diagram of the experimental setup used. Solid lines: scanning measurements of particle mass for determination of extinction cross-sections and mass-mobility scaling exponents and isolation of $q = +1$ particles. Dashed lines: measurements of absorption cross-sections at a static, pre-determined mass setpoint for MM-RIR. Dotted lines: measurements of absorption and extinction coefficients and size distributions for FD-RIR. Abbreviations: differential mobility analyzer (DMA), aerosol particle mass analyzer (APM), cavity ring-down spectrometer (CRD), photoacoustic spectrometer (PA) and condensation particle counter (CPC).

Considering the number of previous investigations that have utilized refractive index retrievals, this is the first study, to our knowledge, performing a comparison of retrieved and calculated values (i.e., inverse and forward comparisons); Bluvshtein et al. (2016) [25] performed a single point comparison of retrieved m and noted that they obtained very good agreement. Single wavelength refractive indices of spherical particles with known composition were retrieved using both full distribution and size- and mass-selected measurements with extinction and absorption being measured by cavity ring-down spectroscopy and photoacoustic spectroscopy, respectively. Particles were generated from ammonium sulfate (AS), nigrosin or a mixture of AS and nigrosin with AS mass fractions (w_{AS}) of 0.75 and 0.50 with multiple distributions being measured for each type to facilitate size-dependent comparisons. Following methods utilized for the retrieval of m from full distribution measurements in other investigations, m was retrieved from: 1) a single retrieval using the set average α_{ext} and α_{abs} and the set average size distribution [31,32,35,37,40,41], 2) multiple retrievals using single measurements of α_{ext} and α_{abs} and the average of 2 distributions (α_{ext} and α_{abs} were measured separately) [25,26,42], and 3) a single retrieval using the set average α_{ext} and α_{abs} and a log-normal fit of the set average size distribution [27–29,33,36,38,39]. We then examine the sensitivity of the set average retrieval (1) by: a) treating the average particle number densities as Poisson distributions, b) "correcting" the measured number densities by the quoted accuracy of the CPC (± 10%) and c) "correcting" the measured the size distributions by shifting them ± 1 size bin (comparable to size corrections for non-spherical particles and refractive index or density dependent sizing) [18,28,34–37,40]. Retrievals were compared by calculating the optical properties measured using the alternate method; e.g., m from MM-RIR were used to calculate the measured optical properties of the full distribution measurements and vice versa. Retrievals were defined as consistent if the calculated values (extinction, absorption and single scattering albedo) agreed with measured values to within 10%. The MM-RIR and Set Average FD-RIR values were also used to calculate the normalized global average aerosol radiative forcing of a model accumulation mode remote continental aerosol to highlight the range of radiative forcing values that can be obtained from small differences in m.

2. Materials & methods

A block diagram of the experimental setup used presently is shown in Fig. 1. Experiments are divided based upon measurement. Solid line: scanning mass distributions to determine C_{ext} for

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