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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 $\approx 15\%$ and $\approx 26\%$ 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.

Keywords

Aerosol; refractive index retrieval; radiative forcing

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

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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)

$$m = n + ik \quad (1)$$

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].

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); Bluvshstein 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 ($\pm 10\%$) 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 Figure 1. Experiments are divided based upon measurement. Solid line: scanning mass distributions to determine C_{ext} for MM-RIR and mass-mobility scaling exponents and to isolate particles bearing $q = +1$. Dashed lines: measurements of C_{abs} at a static, pre-determined mass setpoint (from the scanned mass distributions) for MM-RIR. Dotted lines: measurements of aerosol α_{ext} and α_{abs} and particle size distributions for FD-RIR.

2.1. Aerosol generation and conditioning.

Aerosols were generated from solution in a liquid jet cross-flow atomizer. Of the 2.2 L min⁻¹ of generated flow, ≈ 0.52 L min⁻¹ was sampled for conditioning and measurement. Excess flow was vented in a laboratory snorkel. Aerosols consisting of AS or mixtures of AS and nigrosin were conditioned using a pair of gas drying tubes with a 20:1 parallel flow of dry air ($< 5\%$ relative humidity) and a silica gel diffusion dryer. Pure nigrosin aerosol was conditioned more aggressively by passing the aerosol stream through a silica gel diffusion dryer, a tube furnace set to 300 °C and a second silica gel diffusion dryer to generate a homogeneous distribution of nigrosin particles in mass density (ρ).

2.2. DMA Calibration.

To ensure sizing accuracy, the DMA was calibrated using polystyrene nanospheres with nominal (actual) diameters of 50 (50 ± 2) nm, 60 (57 ± 4) nm, 100 (102 ± 3) nm, 150 (147 ± 3) nm, 200 (203 ± 5) nm, 240 (240 ± 5) nm, 500 (457 ± 10) nm and 700 (701 ± 6) nm. For all measurements, the relative humidity inside the DMA was monitored to ensure it was stable ($< 10\%$) for the duration of an experiment to minimize hygroscopic water uptake and evaporation and/or condensation related interferences from particle bound water in the photoacoustic spectrometer [54–57].

2.3. Cavity ring-down spectrometer.

Extinction measurements by the cavity ring-down spectrometer (CRD) were made using the methods described in Radney and Zangmeister (2016) [58]. Light from a continuous wave diode laser ($\lambda = 660$ nm) pumps a high finesse optical cavity to saturation (mirror reflectivity $> 99.98\%$, transmission $\approx 0.002\%$). The light intensity is rapidly terminated (≈ 10 ns) using an acousto-optic modulator whereby it decays passively and exponentially from mirror losses, gas absorption and aerosol scattering and absorption within the cavity. Aerosol extinction coefficients (α_{ext}) are calculated from the difference between aerosol laden and HEPA-filtered (aerosol free) conditions. Nominal ring-down times for the HEPA-filtered air were 26 μs . CRD saturation occurred at ring-down times < 5 μs corresponding to a maximum α_{ext} of $\approx 5.4 \times 10^{-4} \text{ m}^{-1}$. For MM-RIR measurements, flow through the CRD was maintained at $\approx 0.52 \text{ L min}^{-1}$ by two CPCs operated in parallel at 0.30 L min^{-1} each with $\approx 0.08 \text{ L min}^{-1}$ of clean air backflush. For FD-RIR measurements, the total and backflush flows through the CRD were maintained, except that the flow exiting the CRD was split between a CPC and scanning mobility particle sizer (SMPS; i.e., a tandem DMA and CPC); see Figure 1. These flows were chosen in order to maintain a similar volumetric flow through the conditioning system ($\approx 0.52 \text{ L min}^{-1}$) while allowing for a 10:1 sheath:aerosol flow in the DMA for both MM-RIR and FD-RIR measurements.

2.4. Aerosol mass measurements.

Distributions of aerosol number density (N) and α_{ext} as a function of particle mass (m_p) were measured using an aerosol particle mass analyzer (APM). The APM was set to have a peak ρ of 1.77 g cm^{-3} , 1.5 g cm^{-3} and 1.4 g cm^{-3} for the AS, nigrosin, and AS-nigrosin mixtures, respectively. Distribution scans were conducted using an APM classification parameter (λ_c) of 0.32 [59–61] with each scan lasting 10 min. Data (α_{ext} , N , m_p) was collected at 100 Hz and then averaged to and logged at 1 Hz. The α_{ext} and N as a function of m_p were then fit to Gaussian distributions

$$N = \sum A_{N,q} * \exp\left(\frac{-(m_p - m_{\text{eff},N,q})^2}{2\sigma_{\text{eff},N,q}^2}\right) \quad (2)$$

$$\alpha_{\text{ext}} = \sum A_{\text{ext},q} * \exp\left(\frac{-(m_p - m_{\text{eff,ext},q})^2}{2\sigma_{\text{eff,ext},q}^2}\right) \quad (3)$$

where A , m and σ are the peak value, average mass and distribution width, respectively. The subscripts N , ext , and q correspond to the number concentration, extinction and charge, respectively. The eff subscript has been included to denote that these are effective masses and widths since the APM separates particles based upon their mass-to-charge ratio: $m_{\text{eff}} = m_p/q$ and $\sigma_{\text{eff}} = \sigma_p/q$. Summations have been included to account for the presence of multiply charged ($q > +1$) particles. In this study, N and α_{ext} were fit individually as the particles investigated were nearly spherical with sufficient resolution between successive charge peaks. This is unlike Radney and Zangmeister (2016) [58] where the number density and extinction distributions had to be globally (simultaneously) fit because of significant overlap between peaks of successive charge. Comparing the global and successive fit routines yielded differences between $m_{\text{eff},N,q}$ and $m_{\text{eff,ext},q}$ and $\sigma_{\text{eff},N,q}$ and $\sigma_{\text{eff,ext},q}$ less than a few percent suggesting that either fitting routine would have been appropriate. From these fits, the extinction cross-section (C_{ext}) was calculated from the ratio of the integrals

$$C_{\text{ext}} = \frac{\int \alpha_{\text{ext}}}{\int N} = \frac{A_{\text{ext},q} \sigma_{\text{eff,ext},q}}{A_{N,q} \sigma_{\text{eff},N,q}} \quad (4)$$

for use in MM-RIR. Reported uncertainties in C_{ext} represent the 1σ propagated uncertainty from all fit coefficients with the 1σ , 1 s measurement uncertainty in extinction and number concentration being included in the fits.

From the mass distributions, particle effective mass density (ρ_{eff}) is calculated from

$$\rho_{\text{eff}} = \frac{6m_p}{\pi D_m^3} \quad (5)$$

For spherical particles, the effective density and bulk density should be nearly equal. For a collection of mobility diameters and mass distributions, the mass-mobility scaling exponent (D_{fm}) can be calculated from

$$m_p = k_0 \left(\frac{D_m}{150 \text{ nm}} \right)^{D_{\text{fm}}} \quad (6)$$

where D_m is an arbitrary mobility diameter and k_0 is the mass of particles at a reference mobility diameter (150 nm). The D_{fm} has been widely used as a surrogate for particle fractal dimension [62, 63] (i.e., morphology) since spherical particles will have $D_{\text{fm}} \approx 3$ while fractal aggregates like soot commonly have D_{fm} between 1.8 and 2.2. Note that while D_{fm}

and the fractal dimension of a particle are defined similarly, they are not directly equivalent [64].

2.5. Photoacoustic spectrometer.

The photoacoustic spectrometer (PA) used presently is similar to those described in Radney et al. (2013) [65] and (2014) [66] with slight modifications to data collection and analysis. Briefly, light from a continuous wave diode laser ($\lambda = 660$ nm) was passed in free space to a resonant acoustic cell. Light intensity from the laser was modulated using a mechanical chopper at the cell's resonant frequency; nominally 1.64 kHz in ambient air at 296 K [67]. A power meter was situated at the resonator exit to measure laser power. Chopper, microphone and power meter signals were digitized at 20 kHz for 5 s; microphone acoustic response spans 10 Hz to 10 kHz. The real and imaginary components of the frequency response function of the microphone and power meter relative to the chopper stimulus were calculated using a Fast Fourier transformation (FFT) with a flat top window in 1 s intervals with 5 ms steps between successive FFTs across the entire 100 k Sample data stream. This resulted in 800 total FFT calculations for a 5 s sample; step-size was determined from the maximum rate at which FFTs for the 5 s sample could be processed in real-time. Calculated values at the chopper modulation frequency were retained and averaged with the excess data being discarded. The 5 s data was averaged to 1 min for the calculation of absorption coefficients (α_{abs}) from

$$\alpha_{\text{abs}} = \frac{\sqrt{(x_s - x_{\text{bkg}})^2 + (y_s - y_{\text{bkg}})^2}}{c_c \beta_m R_{\text{PM}} \sqrt{8} \sqrt{x_{\text{pwr}}^2 + y_{\text{pwr}}^2}} \quad (7)$$

where x and y represent the real and imaginary components of the FFT. The subscripts s , bkg and pwr correspond to the sample, background (aerosol free) and power meter signals, respectively. The terms R_{pm} , C_c and β_m represent the transfer function of the power meter, the calculated cell constant of the acoustic resonator and the microphone's sensitivity; presently, $R_{\text{pm}} = 0.709696 \text{ W V}^{-1}$ ($\lambda = 660$ nm) and $C_c \beta_m = 0.187 \text{ V m W}^{-1}$. The $\sqrt{8}$ in Eq. 7 has been included to convert the power meter signal from root mean squared (from the FFT) to peak-to-peak. The 1 min data was averaged to either 5 min or 3 min for the MM-RIR and FD-RIR, respectively. The α_{abs} limit of detection (LOD) was calculated for each data set based upon the 1σ standard deviation of five one min background measurements (see discussion in Section 3.3). These uncertainties were only used to determine the LOD and were not propagated through to the uncertainty in α_{abs} ; α_{abs} uncertainty was taken as the 1σ standard deviation of the one min data points.

For absorption measurements used in MM-RIR, the APM was set to the average m_p from the fit of N versus m_p . Data for each D_m and m_p combination were collected for 5 min. Absorption cross-sections (C_{abs}) were determined from the 1 min averaged α_{abs} and N

$$C_{\text{abs}} = \frac{\alpha_{\text{abs}}}{N} \quad (8)$$

and then averaged to 5 min. The reported uncertainties in C_{abs} represent the 1σ propagated uncertainty from the 1 min averages of α_{abs} and N .

2.6. Mass-mobility refractive index retrieval.

For refractive index retrievals using mobility and mass selected data (MM-RIR), refractive indices were determined by iterating over a range of n and k values with $1.33 < n < 2.5$ and $0 < k < 1$ and finding the minimum of the χ^2 merit function where

$$\chi^2 = \sum \left(\frac{C_{\text{ext, meas}, D_m} - C_{\text{ext, calc}, D_m}}{\sigma_{\text{ext, meas}, D_m}} \right)^2 + \left(\frac{C_{\text{abs, meas}, D_m} - C_{\text{abs, calc}, D_m}}{\sigma_{\text{abs, meas}, D_m}} \right)^2 \quad (9)$$

where the subscripts ext, abs, meas, calc and D_m correspond to values from the extinction, absorption, measured, calculated and at a specific mobility diameter, respectively. The summation has been included to account for the multiple size/mass combinations used in the calculation. Uncertainties in the refractive index were taken to be the locus of points satisfying

$$\chi^2 \leq \chi_{\text{min}}^2 + \chi_{\text{crit}}^2 \quad (10)$$

assuming 2 degrees of freedom (n & k). Refractive indices were calculated with $\chi_{\text{crit}}^2 = 3.677$ corresponding to the upper tail of a 1-sided χ^2 distribution with a cumulative percentile of 84.1 % (i.e., 1σ).

2.6. Full distribution refractive index retrievals.

For the FD-RIR measurements, conditioned aerosol was passed through a dilution stage consisting of a HEPA-filter and orifice placed in parallel causing a majority of the aerosol (95 %) to be filtered. This reduced the aerosol concentration sufficiently such that the same solution concentrations could be used in the aerosol generator for both the MM-RIR and FD-RIR measurements. By using the same solution concentrations, uncertainties arising from differences in morphology (ρ_{eff} and D_{fm}), and hence cross-sections and m , were minimized. The conditioned aerosol stream was passed through the PA and CRD in series and then split to a CPC and a SMPS. Particle size distributions were measured for 180 s and spanned $D_m = 14.6$ nm to 710.5 nm with the 1 min α_{ext} and α_{abs} being averaged to the length of a single scan. During each size distribution measurement, N and either α_{ext} or α_{abs} were measured; extinction and absorption measurements were alternated on successive size distribution scans.

Values of m utilizing FD-RIR were determined by: 1) performing a single FD-RIR using the set average α_{ext} and α_{abs} and set average size distribution for an entire experiment (a.k.a. Set Average FD-RIR) [31, 32, 35, 37, 40, 41], 2) multiple FD-RIR 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], 3) a single FD-RIR using the set average α_{ext} and α_{abs} and a log-normal fit of the set average size distribution for an entire experiment [27–29, 33, 36, 38, 39]. The sensitivity of the Set Average FD-RIR was then investigated by: a) treating the average particle number densities as Poisson distributions, b) “correcting” the measured $N_{\text{by}} \pm 10\%$, the quoted accuracy of the CPC, 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].

Refractive indices were retrieved by iterating over a range of n and k values and finding the minimum of the χ^2 merit function where

$$\chi^2 = \left(\frac{\alpha_{\text{ext, meas}} - \sum N_{D_m} C_{\text{ext, calc}, D_m}}{\sigma_{\text{ext, meas}}} \right)^2 + \left(\frac{\alpha_{\text{abs, meas}} - \sum N_{D_m} C_{\text{abs, calc}, D_m}}{\sigma_{\text{abs, meas}}} \right)^2 \quad (11)$$

The subscripts ext, abs, meas, calc and D_m correspond to values from the extinction, absorption, measured, calculated and at a specific mobility diameter, respectively. The summation has been included to account for the fact that α_{ext} and α_{abs} were calculated for the entire distribution. Similar to the MM-RIR, uncertainties in the refractive index were taken to be the locus of points satisfying Eq. 10 assuming 2 degrees of freedom (n & k); for the individual scan retrievals, the individual χ^2 values were added to obtain a single χ^2 for the entire experiment. A $\chi^2_{\text{crit}} = 3.677$ was used corresponding to the upper tail of a 1-sided χ^2 distribution with cumulative percentile of 84.1 % (i.e., 1σ); for the Poisson – 2σ sensitivity test, $\chi^2_{\text{crit}} = 7.544$ corresponding to a cumulative percentile of 97.7 % (i.e., 2σ).

3. Results and Discussion

Refractive indices and size distributions represent two of the primary input parameters for calculating aerosol optical properties in radiative forcing simulations [1–3]. As a result, refractive index retrievals have been applied to a wide array of atmospheric particles. These retrieval methods require either serial measurements utilizing size- (and mass-) selection to determine the C_{ext} , C_{scat} and/or C_{abs} (or Q_{ext} , Q_{scat} and/or Q_{abs}) or parallel measurements of α_{ext} , α_{scat} and/or α_{abs} and the aerosol size distribution. In this study, particles were generated from AS (a non-absorbing inorganic salt), nigrosin (an absorbing organic dye), and mixtures of AS and nigrosin with AS mass fractions ($w_{\text{AS}} = m_{\text{AS}}/m_{\text{Total}}$) of 0.75 and 0.50. Multiple distributions for each particle type were measured; see Table 1. Individual retrievals were defined as self-consistent if the optical properties measured during data collection (extinction, absorption and albedo) could be calculated from the retrieved m to better than 10 %. Inter-methods comparisons (MM-RIR versus FD-RIR) were defined as consistent if the optical properties measured during data collection for one method could be calculated to better than 10 % using m retrieved by the other method. The sensitivity of the

FD-RIR to the choice of data analysis method and the accuracy of CPC counting and DMA sizing was also investigated. To this end, this section has been organized as follows: §3.1) DMA calibration to ensure accurate measurement of spherical particle diameters, §3.2) effective densities and mass-mobility scaling exponents to demonstrate particle sphericity and the applicability of Mie theory, §3.3) MM-RIR and comparisons to other investigations, §3.4) FD-RIR and sensitivity tests, §3.5) comparison of retrieved refractive indices, §3.6) consistency between retrieval methods and §3.7) implications for radiative forcing calculations. These retrievals were also used to form recommendations for improving refractive index retrievals in future studies.

3.1. DMA Calibration.

The DMA column was cleaned and calibrated using nine monodisperse polystyrene nanosphere aqueous suspensions with nominal diameters ranging from 50 nm to 700 nm; see Table S1 in the Supplementary Materials. To determine the average particle mobility diameter ($D_{m,avg}$), the portion of the distribution corresponding to the monomeric $q = +1$ particle of interest [58, 68] were fit to a log-normal distribution. Particles with actual diameters 100 nm, exhibited relative errors of 2.7 %; Relative error = (Actual diameter – $D_{m,avg}$)/(Actual diameter) $\times$ 100 %. Particle diameters smaller than 100 nm had significantly larger errors and similar to other investigations [69], these differences are attributed to surfactant coatings on the particles that remain as a result of the large droplets generated from a pneumatic atomizer.

3.2. Particle Sphericity.

Both the FD-RIR and MM-RIR methods typically rely upon Mie theory for the retrieval of m . Two of the fundamental assumptions of Mie theory are particle homogeneity and sphericity. In the present investigation, two pure materials (AS and nigrosin) and a pair of mixtures of these materials with $w_{AS} = 0.75$ and 0.50 were investigated; we assume the mixtures form homogeneous particles due to the co-atomization of the material from a mixed aqueous solution.

The assumption of particle sphericity was tested by measuring the average ρ_{eff} and calculating k_0 and D_{fm} (see Table 1, measured m_p versus D_m data is plotted in Figure S1 of the Supplementary Materials) for each w_{AS} and distribution for particles spanning 100 nm D_m 500 nm at 50 nm increments. The geometric mean diameter (D_{geo}) and geometric standard deviation (σ_{geo}) correspond to the average values measured during FD-RIR data collection. Values shown in parenthesis in Table 1 correspond to either the 1σ standard deviation of multiple measurements (D_{geo} , σ_{geo} and ρ_{eff}) or the 1σ fit uncertainty (k_0 and D_{fm}). In fitting k_0 and D_{fm} , the 1σ width of the mass distribution ($\sigma_{eff,N,q}$ in Eq. 2) was treated as the measurement uncertainty; this value is an overestimation of the actual width of the mass distribution due to peak broadening in the APM [60, 61, 70]. The ρ_{eff} exhibit coefficients of variation (CV = uncertainty divided by the average $\times$ 100 %) 4 % at 1σ for $w_{AS} = 1, 0.75$ and 0 indicating that the particles possess a similar morphology independent of particle size and is confirmed by $D_{fm} \approx 3.0$ (within measurement uncertainty) indicating particle sphericity [71–73]. For $w_{AS} = 0.5$, the CV is ≈ 8 % and ρ_{eff} is slightly correlated

with D_m . This is confirmed by $D_{fm} > 3.0$ (outside measurement uncertainty), indicating that for $w_{AS} = 0.5$, particle morphology is slightly D_m dependent.

The ρ_{eff} , k_0 and D_{fm} values between data sets with the same w_{AS} , but different D_{geo} and σ_{geo} , are equivalent within measurement uncertainty across all measured samples (see Table 1). Similarly, the measured C_{ext} and C_{abs} of the different D_{geo} for the same w_{AS} are all within $\pm 10\%$ (See Figure 2 and Table 2). For $w_{AS} = 0.50$, C_{abs} exhibits an average deviation of 20 % between data sets as determined from the slope. By treating C_{abs} for $D_m = 400$ nm as an outlier (see Figure S18 in the Supplementary Materials), this average deviation drops to 10 %. This consistency in ρ_{eff} , k_0 and D_{fm} , C_{ext} and C_{abs} implies that the particles are morphologically and optically similar for a given w_{AS} . Further, the small differences in ρ_{eff} imply that particles within each was should have comparable n [74–76]. While it is unclear if ρ impacts k , it stands to reason that for comparable ρ , C_{ext} and C_{abs} , significant variations in k should not be observed.

These observed similarities can directly (indirectly) affect MM-RIR (FD-RIR) since the cross-sections are directly (indirectly) used in the retrieval; see Equations 9 and 11. For particles that are physically, chemically and morphologically invariant with D_m , this implies that the value of and uncertainty in m obtained from MM-RIR will be nearly independent of the collection of D_m used and decrease with larger collections, respectively [77]. For FD-RIR, the retrieved m is also dependent upon the measured size distribution implying that differences in the retrieved m may be observed even when the particles are physically, chemically and morphologically invariant. As a result of these diagnostic capabilities, the FD-RIR is compared to the MM-RIR, even though the “correct” m for these nanoparticles may not be absolutely known.

3.3. Mass-mobility refractive index retrievals.

Refractive indices from 2 different size distributions of particles composed of AS, nigrosin or AS and nigrosin mixtures with $w_{AS} = 0.75$ or 0.50 were retrieved using particles that were size-selected by a DMA and mass-selected using an APM. The measured data for particles with $w_{AS} = 0$ (nigrosin) and $D_{geo} = 128$ nm is shown as a function of D_m in Figure 3: a) peak α_{ext} (black circles) and N (green triangles) from fits of size distribution scans, b) 5 min average α_{abs} (red squares) and corresponding N (blue diamonds) measured at the average mass ($m_{eff,N,q}$ in Eq. 2, $q = +1$) determined from the mass distribution scans, c) average C_{ext} (black circles) and C_{abs} (red squares). The red line in Figure 3b corresponds to the LOD for this data set (see Figure S2 in the Supplementary Materials and discussion below). The measured data for all remaining samples listed in Table 1 can be found in the Supplementary Materials. Uncertainties have been included in Figure 3 but are not visible as the 1σ measurement uncertainties in the measured data (Figures 3a and 3b) and the propagated uncertainties in cross-sections (Figure 3c) are all smaller than the corresponding data points; The 1σ average CV for α_{ext} , N , and C_{ext} are 1.1 %, 1.3 % and 2.0 %, respectively. For α_{abs} , the 1σ measurement uncertainty is $\approx 2 \times 10^{-5} \text{ m}^{-1}$, independent of absorption since the largest source of uncertainty was microphone self-noise and ambient pick-up noise. As a result, the CV is inversely correlated to α_{abs} with C_{abs} following a similar trend.

An Allan variance [78] was conducted using particle free air to determine the theoretical minimum LOD (see Figure S2 in the Supplementary Materials) and the optimized averaging interval for the 1 min data. From the Allan variance, α_{abs} LOD continuously decreases as $1/\sqrt{N_{\text{samples}}}$ for the entire 30 min interval. As a result, mass-mobility selected absorption data was chosen to be collected for 5 min for which the theoretical LOD was $1.7 \times 10^{-5} \text{ m}^{-1}$. In practice, however, the average background deviation in the microphone signal varied between data sets causing the α_{abs} LOD to range between $1.5 \times 10^{-5} \text{ m}^{-1}$ and $5.5 \times 10^{-5} \text{ m}^{-1}$ (average LOD $\pm 1\sigma = 3.6 \times 10^{-5} \text{ m}^{-1} \pm 1.4 \times 10^{-5} \text{ m}^{-1}$); here, we define the LOD as three times the absorption coefficient calculated from the 1σ standard deviation of the background measurements. For completeness, and to account for the changing LOD, the α_{abs} LOD have been included as a red line in all measured data plots presented in this manuscript and Supplementary Materials. For α_{ext} , the 1 s LOD (defined as three times the standard deviation of the background) was $2.5 \times 10^{-6} \text{ m}^{-1}$, independent of data set.

MM-RIR were performed by iterating over n and k with the value that minimized the χ^2 merit function (Eq. 9) being taken as m for the sample. All data with $D_{\text{m}} > 100 \text{ nm}$ that was above the LOD was included. Although the $D_{\text{m}} = 100 \text{ nm}$ data is shown in most plots of mass- and mobility-selected data, it was not included in the MM-RIR for 2 reasons: 1) the cross-sections are small and require large number concentrations to be above the LOD, even for α_{ext} , so small inaccuracies in the measured coefficients can lead to large errors ($> 100\%$) in the calculated cross-sections [77] – for example, the C_{ext} of a 100 nm AS particle ($n = 1.52$ at $\lambda = 660 \text{ nm}$ [79]) is $1.01 \times 10^{-16} \text{ m}^2$ requiring $\approx 2.5 \times 10^{10}$ particles m^{-3} to obtain $\alpha_{\text{ext}} = 2.5 \times 10^{-6} \text{ m}^{-1}$ (the LOD of the CRD) – and 2) α_{abs} was measured by setting the APM to the average m_{p} determined from scanned distributions of N and α_{ext} . At $D_{\text{m}} = 100 \text{ nm}$, the peak resolution (Rs) [58] between the $q = +1$ and $+2$ particles was ≈ 0.6 for the scanned distributions causing the statically measured α_{abs} at $D_{\text{m}} = 100 \text{ nm}$ to likely contain contributions from $q = +2$ particles due to distribution overlap.

The m retrieved for each x_{AS} and D_{geo} combination are shown in Table 3. Refractive indices for all FD-RIR analysis methods and sensitivity tests have also been included in Table 3, but will be discussed in the following sections. Uncertainties for all retrieved m have been tabulated by algorithm or sensitivity test in Tables S2 through S12 and plotted by w_{AS} and D_{geo} in Figures 6 and S26 through S32. Although the uncertainties are tabulated by method in Tables S2 through S12, the plotted uncertainties in Figures 6 and S26 through S32 are the preferred metric since: 1) we defined the uncertainty as the locus of values that satisfy Eq. 10 which leads to asymmetric uncertainties in both n and k and 2) n and k are not independent of each other but rather complexly coupled (See Bohren and Huffman [4] or Eq. 2.13a and 2.13b of Moosmüller et al. (2009) [80]) leading to regions of uncertainty that cannot be described by a simple $(n \pm u_c(n)) + (k \pm u_c(k))i$ relationship as is typically encountered [81]. This treatment is analogous to Sumlin et al. (2018) [82].

The $x_{\text{AS}} = 1$ retrieved m exhibit good agreement with the bulk AS refractive index ($n = 1.52$ to 1.53 at $\lambda = 660 \text{ nm}$ [79]). Note that only C_{ext} data was used in this retrieval since AS is non-absorbing and the measured α_{abs} for both data sets were below the LOD (measured data can be found in Figures S3 and S6 of the Supplementary Materials). Both $x_{\text{AS}} = 1$

distributions ($D_{\text{geo}} = 113$ nm and 147 nm) exhibited ρ_{eff} less than the crystalline bulk mass density ($\rho = 1.77$ g cm⁻³). Assuming the differences in ρ_{eff} are surface imperfections, void volume inside the particles or slight deviations from sphericity, an effective real component of the refractive index (n_{eff}) can be calculated from the corresponding values for AS (n_{AS}) and air (n_{air}) and their mass fractions (w_{AS} and w_{air} , respectively) [83, 84]

$$n_{\text{eff}} = n_{\text{AS}}w_{\text{AS}} + n_{\text{air}}w_{\text{air}} \quad (12)$$

Applying this correction, n_{eff} is 1.511 to 1.521 and 1.500 to 1.508 for the $D_{\text{geo}} = 113$ nm and 147 nm data sets, respectively, which exhibits significantly better agreement to the retrieved m of $1.513 + 0i$ and $1.504 + 0i$; see Table S2 and Figures S26 and S27 of the Supplementary Materials for uncertainties. Calculated C_{ext} using the retrieved n were self-consistent (see Figure S4 and S7 in the Supplementary Materials) with an average relative deviation ($[(C_{\text{ext,measured}} - C_{\text{ext,calculated}})/C_{\text{ext,measured}}] \times 100\%$) across both $w_{\text{AS}} = 1$ data sets of $< -0.5\%$. This small deviation also serves as a diagnostic ensuring CPC accuracy since C_{ext} is a function of N .

Nigrosin has seen significant use in the literature as a test aerosol because it is water soluble, forms spherical particles and exhibits relatively strong absorption across the UV to near-IR [14, 15, 21, 23, 85–89]. The MM-RIR values for nigrosin with $D_{\text{geo}} = 115$ nm and 128 nm were $1.776 + 0.228i$ and $1.799 + 0.256i$, respectively; see Table S2 and Figures 6 and S32 for uncertainties. Comparing the two nigrosin data sets against each other, ρ_{eff} , C_{abs} and C_{ext} exhibit average differences of $\approx 2\%$, $\approx 1\%$ and $\approx 7\%$, respectively, implying that small differences between the two MM-RIR values should be obtained; see Figure 2 and Table 2. Further, using the retrieved m to calculate C_{ext} , C_{abs} and SSA , the average relative deviations are $< 10\%$ for $D_{\text{m}} > 100$ nm (See Figure 4 and S23) demonstrating that both data sets are self-consistent.

Nigrosin m have been retrieved at multiple λ [15, 16, 21, 32, 85, 90–92]. However, for nigrosin, both n and k are wavelength dependent [21, 85], limiting the applicability of some values in the present investigation. Values for m retrieved at similar λ in the literature were $1.77 + 0.27i$ at $\lambda = 652$ nm [32], $1.7 + 0.24i$ at $\lambda = 670$ nm [91] and $1.70 (\pm 0.02) + 0.28 (\pm 0.01)i$ [21] and $1.812 (\pm 0.0068) + 0.2461 (\pm 0.0031)i$ [85] at $\lambda = 660$. Radney and Zangmeister (2015) [21] measured an average ρ_{eff} for nigrosin of 1.34 g cm⁻³ which is 9 % and 11 % lower than the ρ_{eff} measured for the $D_{\text{geo}} = 115$ and 128 nm data sets, respectively. These differences in ρ_{eff} could account for the differences in the refractive indices relative to previously measured values. Notably, the best agreement to previously reported m are for thin film measurements of nigrosin using spectroscopic ellipsometry [85], a measurement technique more immune to nanoscale density effects [93].

Comparison of measured and calculated values for MM-RIR shows that self-consistency is not always achieved across an entire data set with some of the relative deviations $\approx 25\%$ (See Figures S4, S10, S13, S16, S20 and S23). This is more common for the mixtures ($w_{\text{AS}} = 0.75$ and 0.50) and C_{abs} (and hence SSA). For all data sets, self-consistency is better for distributions with larger D_{geo} which leads to higher particle concentrations and measured

α_{abs} values; hence a lower CV for α_{abs} and C_{abs} . The measured C_{abs} always exhibited larger CVs compared to C_{ext} – average 1σ CV across all data sets were 22 % versus 1 %, respectively – resulting in the C_{ext} data being weighted more heavily in the retrieval (See Eq. 9). Comparing retrievals using just C_{ext} or C_{abs} data yields different m then in combination with the C_{ext} retrieval being closer to the combined value. For example, retrieving m for the $w_{\text{AS}} = 0.5$ and $D_{\text{geo}} = 114$ nm data set (see Figure S18 in the Supplementary Materials) using only C_{ext} or C_{abs} data yields $m = 1.65 + 0.10i$ or $1.73 + 0.17i$, respectively, compared to $1.62 + 0.125i$ using both data sets. Similar to previous observations, the combined data set gives the best agreement with the measured data as a result of the intersecting contours that represent C_{ext} and C_{abs} as a function of m [77, 82]. Importantly, while using both C_{ext} and C_{abs} in the retrieval of m for $w_{\text{AS}} = 0.75$ and 0.50 improves self-consistency, it still may not be achieved (relative deviations $> 10\%$) indicating that either m is D_{m} -specific or a non-homogeneous morphology is being observed requiring the use of more complex optical model (e.g., core-shell); for $w_{\text{AS}} = 0.5$, ρ_{eff} was slightly D_{m} -dependent implying that m likely follows a similar trend. To extend this observation, m retrievals for particles with even more complex morphologies than observed here, such as coated or embedded black carbon particles, could exhibit even less self-consistency. Further exploration of errors in retrievals of m for particles with complex morphologies is beyond the scope of this manuscript.

3.4. Full distribution refractive index retrievals.

Complex refractive indices were retrieved for the eight samples shown in Table 1 from full distribution measurements of α_{ext} , α_{abs} and the corresponding size distributions; retrieved m are shown in Table 3. A representative set of measured data is shown in Figure 5 for nigrosin ($w_{\text{AS}} = 0$, $D_{\text{geo}} = 128$ nm). Measured data plots for the remaining seven data sets can be found in the Supplementary Materials (Figure S5, S8, S11, S14, S17, S21 and S24). Size distributions were collected at 3 min intervals with alternating α_{ext} and α_{abs} measurements. A single diode laser was used for both the PA and CRD instruments and switching between the two measurements was accomplished using a flip optical mount. A CPC and SMPS were operated in parallel downstream of the two spectrometers to maintain a total volumetric flow of 0.6 L min^{-1} and to allow for a comparison of N measured by the CPC and the SMPS. For all measurements, the CPC measured marginally higher than the SMPS which arises from particles at the smallest sizes of the distribution; the minimum diameter counted by the CPC was < 10 nm ($D_{50} = 4$ nm) while the minimum diameter classified by the SMPS scans was 14.6 nm.

In processing the full distribution data, refractive index retrievals were performed using: 1) a single retrieval using the set average α_{ext} and α_{abs} and size distribution for an entire experiment (a.k.a. Set Average FD-RIR) [31, 32, 35, 37, 40, 41], 2) multiple retrievals using single measurements of α_{ext} and α_{abs} and the average of 2 successive 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 for an entire experiment [27–29, 33, 36, 38, 39]. The sensitivity of the Set Average FD-RIR to measured parameters was then investigated by: a) treating the average N as Poisson distributions, b) “correcting” the measured N based upon the prescribed accuracy ($\pm 10\%$) of the CPC, and c) “correcting” the measured size distributions by shifting them ± 1 size bin (comparable to

size corrections for non-spherical particles and m - or ρ -dependent sizing) [18, 28, 34–37, 40]. The corresponding m retrievals and uncertainties can be seen in Figure 6 for nigrosin ($w_{AS} = 0$) with $D_{geo} = 128$ nm. The remaining retrievals and uncertainties can be found by w_{AS} and D_{geo} in Figures S26 through S32 and by algorithm and sensitivity test in Tables S3 through S12.

3.4.1. Single retrieval from set average.—Refractive indices for the eight samples shown in Table 1 were retrieved by averaging across all measured size distributions, α_{ext} and α_{abs} for a given data set; for all samples, charge correction algorithms were applied to the measured size distributions. Retrieved m are shown in the Set Average columns of Table 3; retrieved m with uncertainties are shown in Table S3 and Figures 6 and S26 through S32. For all retrievals, both α_{ext} and α_{abs} were used, regardless of whether α_{abs} was above or below the LOD as using only a single optical value results in a poorly constrained system that can lead to unreasonable values for non-metallic particles (e.g., $n < 1$ or > 2.5 , $k \approx n - 1$ [94] or $k \gg 0$ for non-absorbing aerosols). All retrievals were self-consistent with the measured optical data; however, the uncertainties are significantly larger (≈ 20 and ≈ 5 times greater for n and k , respectively) than the MM-RIR values. These values were also computed utilizing the contour intersection method of PyMieScatt; see of Sumlin et al. (2018) [82]. Excellent agreement between the two methods (contour intersection versus χ^2 minimization) was obtained with average n and k less than 0.002 and 0.001, respectively, for all retrievals. Comparison of the χ^2 minimization to the survey iteration yielded similar results except that an m for the non-absorbing AS could not be obtained.

3.4.2. Multiple retrievals from size distributions.—As a comparison to the single m retrieval using an average size distribution, m were retrieved for pairwise combinations of full distribution measurements. This corresponds to a single 3 min measurement of α_{ext} and α_{abs} and the average of two size distributions; a single diode laser was used for both the PA and CRD, where α_{ext} and α_{abs} were measured serially. Values of χ^2 were calculated for each measurement by iterating over n and k following Eq. 11. The collection of χ^2 values for the entire data set were then summed with m being taken as the value with the minimum aggregated χ^2 from the multiple retrievals with the uncertainty being defined as in Eq. 10. Alternatively, an m for each retrieval was determined from the minimum χ^2 with the m and uncertainty of the data set being calculated from the average and the standard deviation of the multiple retrievals (this data is shown in green in the Multiple panel of Figures 6 and S26 through S32). The m from these two retrievals are listed as Multiple and Mean, respectively, in Table 3. Uncertainties are tabulated in Tables S4 and S5, respectively, and plotted in Figures 6 and S26 through S32. In general, agreement with Set Average FD-RIR is excellent with average n and k of 0.003 and -0.001 and 0.003 and -0.002 for the Multiple and Mean algorithms, respectively. While the values of n and k agree rather well, the differences in uncertainties are significant; the uncertainty in n and k for the Multiple algorithm are 70 % smaller and 70 % larger than the Set Average algorithm, respectively. This difference arises because the uncertainty in α_{abs} is larger for the individual retrievals than the average while the opposite is true for α_{ext} ; the CRD, and hence α_{ext} , is significantly more sensitive to small perturbations in N or the size distribution than the PA and α_{abs} . The Mean algorithm has the lowest combined uncertainties of all algorithms investigated since the

uncertainties were calculated from multiple retrievals and not the 2-dimensional n and k space that satisfy Eq. 10. Thus, while these values give a measure of the variability in the retrieved values between scans, they do not necessarily give a measure of the allowable variability in optical parameterization.

3.4.3. Single retrieval from log-normal fit.—Full distribution m were also retrieved using a monomodal log-normal fit of the average size distributions with the assumption that the size distributions were well described by a single log-normal distribution [95]; the recalculated m are listed under Fit in Table 3 with m and uncertainties tabulated in S6 and plotted in Figures 6 and S26 through S32. The log-normal fits resulted in a decrease in the retrieved refractive index – average absolute n and k are 0.054 and 0.010 relative to the Set Average (calculated as Set Average – Fit), respectively, for all data sets that arises because the “tail” of the size distribution at large D_m decreases faster in the measured than the fitted data. Compared to the MM-RIR average absolute n and k are 0.085 and 0.032, respectively. The retrieved m for the fitted data are always less than the Set Average and MM-RIR since the fit includes a greater N at large D_m than measured, with these individual particles exerting a larger fractional contribution to α_{ext} and α_{abs} (larger C_{ext} and C_{abs} , respectively).

3.4.4. Poisson counting statistics.—To test the sensitivity of the Set Average FD-RIR to measured N near 0 m^{-3} (i.e., low N at larger D_m where count significance is questionable), the average N at each D_m was treated as a Poisson distribution [96, 97]. The CPC counts c particles per unit volume V giving $\lambda_{\text{poi}} = c/V$ and the probability distribution $\lambda_{\text{poi}} V$ such that

$$P(N) = \frac{e^{-\lambda_{\text{poi}} V} (\lambda_{\text{poi}} V)^c}{c!} \quad (13)$$

Therefore, the average N and approximate uncertainty take the form [96]

$$N \pm T\sqrt{N} \quad (14)$$

where T is the left-tailed t -statistic for a given cumulative percentile and two degrees of freedom; at 1σ and 2σ , $P = 84.1\%$ and $T = 1.32$ and $P = 97.7\%$ and $T = 4.50$, respectively. At D_m with N that were not statistically significant (i.e., $N - T\sqrt{N} \leq 0$), N was set to 0, and m recalculated; retrieved m are shown in the Poisson – 1σ and Poisson – 2σ columns of Table 3 with uncertainties tabulated in Table S7 and S8 and plotted in Figures 6 and S26 through S32. Agreement of m with the MM-RIR improves slightly with this treatment. Average n and k relative to the MM-RIR for the 1σ and 2σ treatments are 0.027 and 0.021 and 0.009 and 0.019, respectively. For comparison, the average n and k for the set average relative to the MM-RIR are 0.031 and 0.022, respectively. Last, deviations in n and k from treating the size distributions with Poisson counting statistics were essentially

independent of w_{AS} and D_{geo} and always improved agreement with the MM-RIR values with the 2σ treatment being better than 1σ .

3.4.5. CPC N accuracy.—To test the sensitivity of the FD-RIR to the CPC's accuracy when $N \gg 0 \text{ m}^{-3}$ (i.e., large N near D_{geo}), measured size distributions were scaled by the quoted accuracy of the CPC; $\pm 10\%$. The retrieved m are listed as CPC – 0.9 and CPC – 1.1 in Table 3 for the plus and minus 10 % cases, respectively. This scaling resulted in an average n and k relative to Set Average FD-RIR of -0.042 and -0.010 and 0.035 and 0.008 for the negative and positive directions, respectively. These shifts in the calculated m , and the corresponding uncertainties, can be found in Tables S9 and S10 and plotted in Figures 6 and S26 through S32. Deviations in n and k arising from the accuracy of N were correlated with k ; larger k exhibited greater n and k .

3.4.6. CPC D_m accuracy.—To test the sensitivity of the retrieved m to the sizing accuracy of the DMA, individual size bins from the Set Average distribution were shifted by minus or plus one bin. Retrieved m are listed under D_m Minus and D_m Plus in Table 3 with uncertainties in Table S11 and S12 and plotted in Figures 6 and S26 through S32. At $D_m = 101.8 \text{ nm}$ with $64 \text{ bins decade}^{-1}$, this corresponds to D_m being shifted to 98.2 nm or 105.5 nm , respectively. For $x_{AS} = 0$ and $D_{geo} = 128 \text{ nm}$, see Figure S25 in the Supplementary Materials, these D_m shifts change D_{geo} to 124 nm and 133 nm and the retrieved m from $1.79 + 0.20i$ to $1.89 + 0.23i$ and $1.71 + 0.18i$, respectively. Similar deviations are observed when performing the D_m minus and plus shifts on the remaining data sets, see Figures 6 and S26 through S32. The average n and k relative to the set average FD-RIR are -0.022 and -0.03 and 0.009 and 0.019 for the minus and plus directions, respectively, with the magnitude of the deviations being correlated with the magnitude of k . Importantly, these D_m shifts loosely correspond to the measured uncertainty (-3.3% to $+3.0\%$) in the determination of particle diameters for 100 nm polystyrene spheres using an uncalibrated DMA [98, 99] and are less than the shifts often applied for DMA shape factor corrections in measurements of non-spherical particles [36], optical particle counter corrections for m -dependent sizing [18, 28, 35, 37, 40] and aerodynamic particle sizer corrections for materials with $\rho \sim 1 \text{ g cm}^{-3}$ [34].

3.5. Comparison of retrieval methods.

A principal goal of this study was to compare the m retrieved using data collected with size- and mass-selected particles to those using the full distributions. Retrieved n and k with uncertainties are shown in Figure 7 for the MM-RIR and Set Average FD-RIR. The FD-RIR consistently returns smaller values of n when compared to the MM-RIR. For absorbing aerosols, k are lower for the Set Average FD-RIR than the MM-RIR by an average of 0.03 units. For non-absorbing aerosols, the FD-RIR consistently returned non-zero k , even though the measured absorption coefficients were not statistically significant. Lastly, the uncertainties associated with FD-RIR were always significantly larger (≈ 22 and ≈ 6 times greater for n and k , respectively) when compared to MM-RIR and is partially attributable to the MM-RIR utilizing at least 7 data points (extinction only for $D_m > 100 \text{ nm}$), but typically more; these ratios are independent of whether a single retrieval from the Set Average or multiple retrievals across a data set were utilized.

Comparing the algorithms and sensitivity tests applied to the FD-RIR in this study reveals a few trends: 1) deviations in the retrieved n (compared to the Set average FD-RIR) are less sensitive to the absolute accuracy of N than D_m (average $n = -0.042$ versus -0.053 for negative deviations and 0.035 versus 0.060 for positive deviations, respectively) while deviations in k are approximately equally sensitive to the absolute accuracy of N and D_m (average $k = -0.010$ versus -0.008 and 0.008 versus 0.011 , respectively). 2) The magnitude of the uncertainties in the retrieved m are independent of the accuracy of N and D_m . 3) Deviations in the retrieved m are correlated with k when the accuracy of either D_m or N is suspect. This implies that larger deviations in m from the “true” value can be expected when using FD-RIR for strongly absorbing aerosols with ill-defined diameters; e.g., soot or black carbon.

3.6. Inter-retrieval consistency.

Using m from MM-RIR, a_{ext} , a_{abs} and SSA were calculated for the size distributions used in the Set Average FD-RIR (see Table 4). Conversely, m from the Set Average FD-RIR were used to calculate C_{ext} , C_{abs} and SSA for the size and mass selected data; these values are shown in red in Figures 4, S4, S7, S10, S13, S16, S20 and S23. The average magnitude of the relative deviation between the measured optical properties for the FD-RIR data using m from MM-RIR are 8.8 %, 21 % and 13 % in a_{ext} , a_{abs} and SSA , respectively. Comparing just the pure materials ($w_{\text{AS}} = 0$ and 1) lowers the deviations to 7.4 %, 17 % and 8.3 %, respectively. Conversely, the average magnitude of the relative deviations between measured C_{ext} , C_{abs} and SSA and those calculated using m from the Set Average FD-RIR are 9 %, 22 % and 48 %, respectively. Comparing just the pure materials (i.e., $w_{\text{AS}} = 0$ and 1) lowers the deviations to 5.6 %, 16 % and 7.6 %, respectively. This decrease in deviations when only examining pure materials points towards the fact that the mixtures may not exist as homogeneous mixtures (which is assumed) but rather something more complicated that requires the use of a more complex optical model to accurately describe the measured values; note, the FD-RIR by itself would not be able to capture these complexities if they are present.

3.6. Implications for radiative forcing calculations.

Particle size distributions and m are the two primary input parameters for determining aerosol optical properties radiative forcing simulations. Simple analytical expressions for the global average aerosol radiative forcing (F_{aer}) have been developed and widely used as a simple method to determine the influence of particle optics and microphysics. One of the more popular variants [9, 27, 100–104] is based upon an expression originally proposed by Chýlek and Wong (1995) [105] or Haywood and Shine (1995) [106] that normalizes F_{aer} to the aerosol optical depth (τ)

$$\frac{\Delta F_{\text{aer}}}{\tau} = \frac{S_0}{2} T_{\text{atm}}^2 (1 - A_{\text{cld}}) [\beta(SSA)(1 - R_{\text{surf}})^2 - 2(1 - SSA)R_{\text{surf}}] \quad (15)$$

where S_0 , T_{atm} , A_{cld} , β and R_{surf} are the solar constant (1370 W m^{-2}), the atmospheric transmittance above the aerosol layer (0.76), the fractional cloud cover (0.6), the upscatter

fraction of the earth's sunlit hemisphere and the surface reflectance (0.15, appropriate for an urban area). The $\frac{1}{2}$ term with S_0 has been included to account for the fractional day length. Using Mie theory with m from the MM-RIR and the Set Average FD-RIR and size distributions corresponding to the accumulation mode (Mode II) of a remote continental aerosol [95, 107] with $N = 2.9 \times 10^9 \text{ m}^{-3}$, $D_{\text{geo}} = 116 \text{ nm}$ and $\sigma_{\text{geo}} = 1.648$, a_{ext} , a_{abs} , a_{scat} and a_{bscat} were calculated with *SSA* and hemispheric backscatter fraction ($b = a_{\text{bscat}}/a_{\text{scat}}$) determined from the calculated values. The upscatter fraction (β) was computed using the fifth order polynomial fit of Moosmüller and Ogren (2017) [100]

$$\beta = 99.69b^5 - 144.6b^4 + 80.44b^3 - 22.05b^2 + 3.73b + 0.018 \quad (16)$$

Calculated F_{aer}/τ values (Table 5) exhibit a wide range of deviations between the Set Average FD-RIR and MM-RIR methods that span from good ($< 10\%$) to poor ($> 100\%$). The calculated F_{aer}/τ values for $w_{\text{AS}} = 0.75$ and $D_{\text{geo}} = 118 \text{ nm}$ and $w_{\text{AS}} = 0.50$ and $D_{\text{geo}} = 132 \text{ nm}$ are even predicted to exhibit different directions of radiative forcing (positive versus negative). These differences in F_{aer}/τ highlight the potential sensitivity of radiative forcing calculations to the input m ; for example, for $w_{\text{AS}} = 0.75$ and $D_{\text{geo}} = 128 \text{ nm}$, n and k are -0.001 and 0.03 , respectively, and yet a deviation of -286% in F_{aer}/τ is obtained.

4. Summary and Conclusions.

This investigation has focused on comparing aerosol complex refractive indices retrieved using measurements of absorption and extinction cross-sections of mobility- and mass-selected aerosols to those retrieved using measurements of absorption and extinction coefficients of full aerosol distributions. In an absolute sense, the complex refractive indices retrieved by the methods sometimes agree rather well (n and $k < 0.02$) but sometimes don't (n and $k > 0.03$) independent of whether the aerosol particles consisted of a pure material or mixture with the Set Average FD-RIR always yielding better consistency than the Fit FD-RIR. Consistency between methods, and occasionally self-consistency, to calculate all the measured optical properties (extinction, absorption and single scattering albedo) from the retrieved complex refractive index to $\sim 10\%$ was rarely achieved; typically, agreement of at least 1 of the measured values to better than 10% was possible. Importantly, the systems studied used: 1) spherical particles with diameters less than $1 \mu\text{m}$ thereby removing the need for size, morphology and/or refractive index dependent correction factors, 2) the same "stock" of aerosols for both the full distribution and mass-mobility selected measurements allowing for direct comparison of the two retrievals and 3) multiple geometric mean diameters for each ammonium sulfate mass fraction allowing for size-dependent comparisons independent of particle chemistry.

In closing, these data may offer some insight into improving the determination of aerosol refractive indices in future studies. Refractive index retrievals utilizing spectroscopic measurements of mobility-selected particles (and mass if available) will most likely remain limited to laboratory measurements where high number densities of aerosol and long-term source stability can be achieved. Full distribution measurements are more likely to be utilized in field measurements where the number densities and physical sizes of particles

encountered are lower and temporally varying [22]. For these retrievals, shorter time resolution (< 1 h) is necessary to track particle evolution.

Using the presented data as a guide, it is recommended that the workup for full distribution refractive index retrievals include some test of count significance at low number densities and use the measured size distributions instead of log-normal fits of the size distribution. Further, reported values for full distribution refractive index retrievals should include size distribution information, as the retrieved refractive index can be a function of the measured distribution independent of particle chemistry; importantly, separating these contributions requires some *a priori* knowledge about the measured particles that is not typically available from optical measurements alone. For mass-mobility refractive index retrievals, self-consistency between measured and calculated cross-sections needs to be ensured since disagreement may necessitate the use of a more complex optical model. While self-consistency is a readily available diagnostic for the mass-mobility refractive index retrievals, full distribution retrievals should always be self-consistent since, in principle, virtually any combination of optical coefficients and size distributions can be equated if the complex refractive index is allowed to vary over a wide enough range. In some instances, the full distribution retrieved refractive index may not be physically reasonable for an atmospheric aerosol (e.g., $k - n - 1$ corresponding to metallic particles [94]). It is possible that some combination of both full distribution and mass-mobility retrieval methods would give an optimal estimation of the complex refractive index. Last, considering of the range of complex refractive index values observed for the simple single- and two-component sub-micron spherical aerosols measured in this study, it should also be recommended that retrieved refractive indices, regardless of the method, only be treated as semi-quantitative since calculated optical properties and radiative forcings can deviate by $> 20\%$ and $> 100\%$, respectively. Notably, significant deviations in the calculated radiative forcing can arise from errors in either the real or imaginary component of the refractive index as small as 0.02. It is likely that these errors are greater for particles that are super-micron, multi-component, non-spherical, possess a complex morphology (e.g., core-shell, fully embedded, partially embedded, etc.) or otherwise violate the assumptions of Mie theory in some way.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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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})
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 $\approx 68.2\%$ 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

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Figure 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).

Figure 2:

Mobility and mass-selected aerosol comparison of measured extinction (C_{ext} , circles) and absorption (C_{abs} , squares) cross-sections as a function of ammonium sulfate mass fraction (w_{AS}) for the pairwise small and large distributions determined from D_{geo} (e.g., common w_{AS} , See Table 1). Solid black line and grey shading correspond to 1:1 and $\pm 10\%$. Uncertainties represent the 1σ propagated measurement uncertainties.

Figure 3:

Measured data used for mass-mobility refractive index retrievals for nigrosin particles ($w_{AS} = 0$) and $D_{geo} = 128$ nm as a function of mobility diameter (D_m). a) Peak fitted extinction coefficients (α_{ext} , black circles, left axis) and number densities (N , green triangles, right axis) from mass distribution scans. b) Measured absorption coefficients (α_{abs} , red squares, left axis) and N (blue diamonds, right axis). Shaded red line represents the limit of detection for this set of absorption measurements. c) extinction (C_{ext} , black circles) and absorption (C_{abs} , red squares) cross-sections calculated from data in a) and b).

Figure 4:

Measured versus calculated a) extinction cross-sections (C_{ext}), b) absorption cross-sections (C_{abs}) and c) single scattering albedos (SSA) and relative deviations – b), d), and f) respectively – for nigrosin particles ($w_{AS} = 0$) with $D_{geo} = 128$ nm as a function of mobility diameter (D_m). Values shown as red squares and green triangles were calculated using complex refractive indices retrieved using the MM-RIR and set average FD-RIR to test for self-consistency and inter-method consistency, respectively. Shaded gray areas in b), d) and f) correspond to $\pm 10\%$.

Figure 5:

Measured data for nigrosin particles ($w_{\text{AS}} = 0$, $D_{\text{geo}} = 128 \text{ nm}$) as a function of SMPS scan (averages shown at far right) used for full distribution refractive index retrievals. a) extinction (α_{ext} , black circles) and absorption (α_{abs} , red squares) coefficients. b) number densities (N) measured by the CPC during extinction measurements (black circles) and absorption measurements (red squares) and from the integrated particle concentrations measured by the SMPS (green triangles). c) Geometric mean diameter (D_{geo}) and d) geometric standard deviation (σ_{geo}) of the size distribution measurements.

Figure 6:

Plots of retrieved complex refractive indices (red squares) and corresponding uncertainties (shaded black) by algorithm and sensitivity test for nigrosin ($w_{AS} = 0$) with $D_{geo} = 128$ nm. See discussions in text.

Figure 7:

Comparison of the real (top) and imaginary (bottom) components of the refractive index across all measured samples retrieved using the Mass-Mobility (MM-RIR) and the set average full distribution refractive index retrieval methods. Grey lines correspond to 1:1. Error bars represent the 1σ retrieval uncertainties, see discussion in text.

Table 1:

Average geometric mean diameter (D_{geo}), average geometric standard deviation (σ_{geo}), average effective mass density (ρ_{eff}), mass-mobility prefactor (k_0), and mass-mobility scaling exponent (D_{fm}) as a function of AS mass fraction (w_{AS}).

w_{AS}	D_{geo} (nm) ^a	σ_{geo} ^a	ρ_{eff} (g cm ⁻³) ^a	k_0 (fg) ^b	D_{fm} ^b
1	113 (2)	1.65 (0.02)	1.74 (0.07)	3.0 (0.3)	3.1 (0.1)
1	147 (2)	1.74 (0.05)	1.72 (0.06)	2.9 (0.3)	3.1 (0.1)
0.75	118 (1)	1.68 (0.02)	1.41 (0.03)	2.5 (0.3)	3.0 (0.1)
0.75	140 (2)	1.70 (0.04)	1.43 (0.03)	2.5 (0.3)	3.0 (0.1)
0.5	114 (1)	1.66 (0.02)	1.5 (0.1)	2.4 (0.3)	3.1 (0.1)
0.5	132 (1)	1.69 (0.04)	1.5 (0.1)	2.4 (0.3)	3.1 (0.1)
0	115 (3)	1.69 (0.03)	1.47 (0.05)	2.5 (0.3)	3.1 (0.2)
0	128 (2)	1.74 (0.03)	1.50 (0.06)	2.6 (0.3)	3.1 (0.1)

^aValues in parenthesis are 1 σ standard deviations of multiple measurements.

^bValues in parenthesis are 1 σ fit uncertainties from Eq. 6.

Table 2:

Slopes of linear best fits for data shown in Figure 2 as a function of mobility and mass-selected AS mass fraction (w_{AS}).

w_{AS}	C_{abs}	C_{ext}
1	--	1.030 (0.003)
0.75	0.92 (0.11)	0.915 (0.002)
0.50	1.20 (0.04)	0.948 (0.003)
0	1.01 (0.01)	0.929 (0.002)

Table 3:

Retrieved complex refractive indices as a function of the mass fraction ammonium sulfate (was) and the measured geometric mean diameter (D_{geo}) using measurements of mass- and mobility-selected particles (MM-RIR) and full distributions (FD-RIR); see discussion in text. Precision of refractive indices reflects their absolute uncertainties; see discussion in text and Tables S2 through S12 and Figures 6 and S26 through S32.

w_{AS}	D_{geo} (nm)	FD-RIR				Fit	Poisson - 1σ	Poisson - 2σ	CPC - 0.9	CPC - 1.1	D_{m} Minus	D_{m} Plus
		Set Average	Multiple	Mean	MM-RIR							
1	113 (2) ^a	1.48 + 0.012 <i>i</i>	1.48 + 0.01 <i>i</i>	1.48 + 0.012 <i>i</i>	1.45 + 0.011 <i>i</i>	1.50 + 0.012 <i>i</i>	1.52 + 0.01 <i>i</i>	1.51 + 0.013 <i>i</i>	1.46 + 0.011 <i>i</i>	1.46 + 0.011 <i>i</i>	1.53 + 0.013 <i>i</i>	1.44 + 0.011 <i>i</i>
1	147 (2) ^a	1.48 + 0.002 <i>i</i>	1.48 + 0.002 <i>i</i>	1.477 + 0.002 <i>i</i>	1.45 + 0.002 <i>i</i>	1.48 + 0.002 <i>i</i>	1.48 + 0.002 <i>i</i>	1.51 + 0.002 <i>i</i>	1.46 + 0.002 <i>i</i>	1.46 + 0.002 <i>i</i>	1.53 + 0.002 <i>i</i>	1.44 + 0.002 <i>i</i>
0.75	118 (1) ^a	1.46 + 0.07 <i>i</i>	1.46 + 0.07 <i>i</i>	1.46 + 0.067 <i>i</i>	1.48 + 0.07 <i>i</i>	1.46 + 0.07 <i>i</i>	1.48 + 0.07 <i>i</i>	1.49 + 0.07 <i>i</i>	1.43 + 0.06 <i>i</i>	1.43 + 0.06 <i>i</i>	1.50 + 0.07 <i>i</i>	1.42 + 0.06 <i>i</i>
0.75	140 (2) ^a	1.52 + 0.066 <i>i</i>	1.52 + 0.04 <i>i</i>	1.522 + 0.036 <i>i</i>	1.511 + 0.035 <i>i</i>	1.521 + 0.036 <i>i</i>	1.53 + 0.036 <i>i</i>	1.55 + 0.04 <i>i</i>	1.492 + 0.033 <i>i</i>	1.492 + 0.033 <i>i</i>	1.57 + 0.04 <i>i</i>	1.476 + 0.033 <i>i</i>
0.50	114 (1) ^a	1.62 + 0.125 <i>i</i>	1.56 + 0.13 <i>i</i>	1.56 + 0.132 <i>i</i>	1.51 + 0.12 <i>i</i>	1.57 + 0.13 <i>i</i>	1.60 + 0.14 <i>i</i>	1.60 + 0.15 <i>i</i>	1.52 + 0.12 <i>i</i>	1.52 + 0.12 <i>i</i>	1.56 + 0.13 <i>i</i>	1.46 + 0.11 <i>i</i>
0.50	132 (1) ^a	1.64 + 0.124 <i>i</i>	1.63 + 0.06 <i>i</i>	1.628 + 0.057 <i>i</i>	1.57 + 0.053 <i>i</i>	1.63 + 0.058 <i>i</i>	1.64 + 0.059 <i>i</i>	1.67 + 0.064 <i>i</i>	1.59 + 0.054 <i>i</i>	1.59 + 0.054 <i>i</i>	1.69 + 0.064 <i>i</i>	1.57 + 0.053 <i>i</i>
0	115 (2) ^a	1.776 + 0.228 <i>i</i>	1.71 + 0.18 <i>i</i>	1.70 + 0.18 <i>i</i>	1.56 + 0.14 <i>i</i>	1.72 + 0.18 <i>i</i>	1.75 + 0.19 <i>i</i>	1.77 + 0.20 <i>i</i>	1.67 + 0.16 <i>i</i>	1.67 + 0.16 <i>i</i>	1.80 + 0.20 <i>i</i>	1.64 + 0.16 <i>i</i>
0	128 (2) ^a	1.799 + 0.256 <i>i</i>	1.79 + 0.20 <i>i</i>	1.79 + 0.20 <i>i</i>	1.67 + 0.17 <i>i</i>	1.79 + 0.20 <i>i</i>	1.81 + 0.21 <i>i</i>	1.87 + 0.23 <i>i</i>	1.73 + 0.18 <i>i</i>	1.73 + 0.18 <i>i</i>	1.89 + 0.23 <i>i</i>	1.71 + 0.18 <i>i</i>

^aValues in parenthesis represent the 1σ standard deviation of D_{geo} for all measured size distributions.

Table 4:Comparison of α_{ext} , α_{abs} and SSA for the set average FD-RIR measured distributions using MM-RIR values

w_{AS}	D_{geo} (nm)	$\alpha_{\text{ext,measured}}$ ($\times 10^{-4} \text{ m}^{-1}$)	$\alpha_{\text{ext,calculated}}$ ($\times 10^{-4} \text{ m}^{-1}$)	Deviation ^a (%)	$\alpha_{\text{abs,measured}}$ ($\times 10^{-4} \text{ m}^{-1}$)	$\alpha_{\text{abs,calculated}}$ ($\times 10^{-4} \text{ m}^{-1}$)	Deviation ^a (%)	SSA_{measured}	$SSA_{\text{calculated}}$	Deviation ^a (%)
1	113 (2)	0.64 (0.05)	0.66	-3.4	--	--	--	--	--	--
1	147 (2)	4.6 (0.5)	4.90	-7.5	--	--	--	--	--	--
0.75	118 (1)	1.9 (0.1)	2.10	-11	0.75 (0.08)	0.73	2.5	0.61 (0.07)	0.66	-7.7
0.75	140 (2)	3.67 (0.05)	4.00	-8.3	0.73 (0.03)	1.25	-72	0.80 (0.03)	0.69	14
0.50	114 (1)	1.82 (0.04)	2.00	-9.6	0.94 (0.06)	0.91	3.1	0.48 (0.03)	0.54	-12
0.50	132 (1)	4.0 (0.1)	4.70	-16	1.01 (0.02)	1.91	-90	0.75 (0.03)	0.59	21
0	115 (3)	2.0 (0.2)	2.40	-16	1.01 (0.05)	1.25	-23	0.51 (0.06)	0.48	5.8
0	128 (2)	4.0 (0.3)	4.20	-5.0	1.82 (0.07)	2.14	-16	0.54 (0.05)	0.49	9.0

^a Deviation = (Measured-Calculated)/Measured * 100%

Table 5:

Calculated model values and optical depth normalized radiative forcing (F_{aer}/τ) computed using retrieved refractive indices.

w_{AS}	D_{geo} (nm)	Method	n	k	$\alpha_{\text{ext}} (\times 10^{-4} \text{ m}^{-1})$	$\alpha_{\text{abs}} (\times 10^{-4} \text{ m}^{-1})$	$\alpha_{\text{scat}} (\times 10^{-4} \text{ m}^{-1})$	$\alpha_{\text{bscat}} (\times 10^{-4} \text{ m}^{-1})$	SSA	b	θ	$F_{\text{aer}}/\tau (\text{W m}^{-2})$	Deviation ^a (W m^{-2})	Deviation ^b (%)
1	113 (2)	FD-RIR	1.482	0.012	13.4	1.3	12.1	2.7	0.9	0.22	0.34	-30.7	-14.1	32
		MM-RIR	1.513	0	14.1	0	14.1	4.2	1	0.29	0.39	-44.8		
1	147 (2)	FD-RIR	1.483	0.002	12.7	0.2	12.5	3.2	0.98	0.26	0.37	-40.3	-3.8	9
		MM-RIR	1.504	0	13.6	0	13.6	3.9	1	0.29	0.39	-44.1		
0.75	118 (1)	FD-RIR	1.456	0.065	16	6.4	9.6	1.2	0.6	0.12	0.26	0.8	-5.1	119
		MM-RIR	1.50	0.062	17.7	6.2	11.5	1.7	0.65	0.14	0.28	-4.3		
0.75	140 (2)	FD-RIR	1.521	0.036	16.9	3.8	13.1	2.5	0.78	0.19	0.32	-17.7	13.1	-286
		MM-RIR	1.52	0.066	18.9	6.6	12.3	1.8	0.65	0.15	0.29	-4.6		
0.50	114 (1)	FD-RIR	1.557	0.133	24.7	12.3	12.4	1.5	0.5	0.12	0.26	8.5	-5.6	-199
		MM-RIR	1.624	0.125	27.2	12	15.2	2.2	0.56	0.14	0.28	2.8		
0.50	132 (1)	FD-RIR	1.625	0.058	23.5	6.2	17.3	3.6	0.74	0.21	0.33	-15.2	17.0	989
		MM-RIR	1.64	0.124	27.9	11.9	15.9	2.4	0.57	0.15	0.29	1.7		
0	106 (3)	FD-RIR	1.713	0.177	33.8	16.2	17.6	2.7	0.52	0.15	0.29	5.5	2.0	27
		MM-RIR	1.776	0.228	38.6	19.7	18.9	3.1	0.49	0.16	0.3	7.6		
0	128 (2)	FD-RIR	1.791	0.203	38.3	18.3	20	3.3	0.52	0.17	0.3	4.7	3.9	45
		MM-RIR	1.799	0.256	40.7	21.4	19.2	3.3	0.47	0.17	0.3	8.6		

^a Deviation = (F_{aer}/τ) - Set Average FD-RIR

^b Deviation = (MM-RIR - Set Average FD-RIR)/MM-RIR * 100 %