Evaluation of Combined Electrical-Mobility and Optical Sizing Techniques for Deriving Aerosol Refractive Index

Stephen Zimmerman, Richard H. Moore, Bruce Anderson, Andreas Beyersdorf, Chelsea Corr, Lee Thornhill, Edward Winstead, and Luke Ziemba

NASA Langley Research Center, Hampton, Virginia, USA

stephenzimm.is@gmail.com, richard.h.moore@nasa.gov

Background and Motivation

- The next generation of aerosol satellite instruments will include multi-spectral polarimetric measurements to retrieval aerosol size distribution and refractive index (Hasekamp et al., 2011; NRC, 2007).
- Refractive index is the "only means of constraining aerosol chemical composition from space" for passive sensors (Mischenko et al., 2007).
- A number of methods have been developed to derive refractive index from combined optical and electrical mobility sizing instruments (e.g., the "Alignment Method" of Hand and Kreidenweis, 2002).
- In this work, we evaluate the sensitivity of two, commercially-available, high-resolution optical particle counters for determining the size-resolved refractive index of laboratory-generated aerosols when used with a modified form of the Alignment Method.

Experimental

The experimental setup is shown below at left:

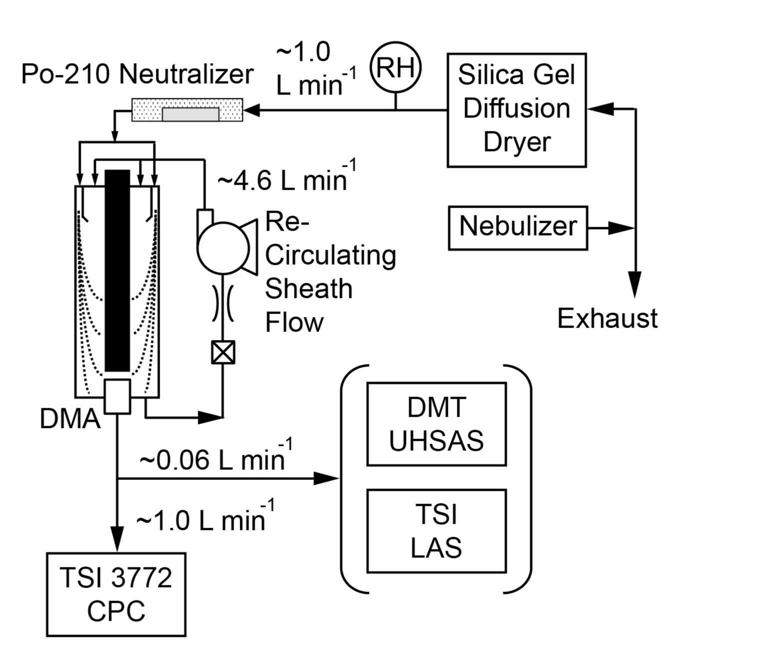


Figure 1. Schematic of the experimental setup.

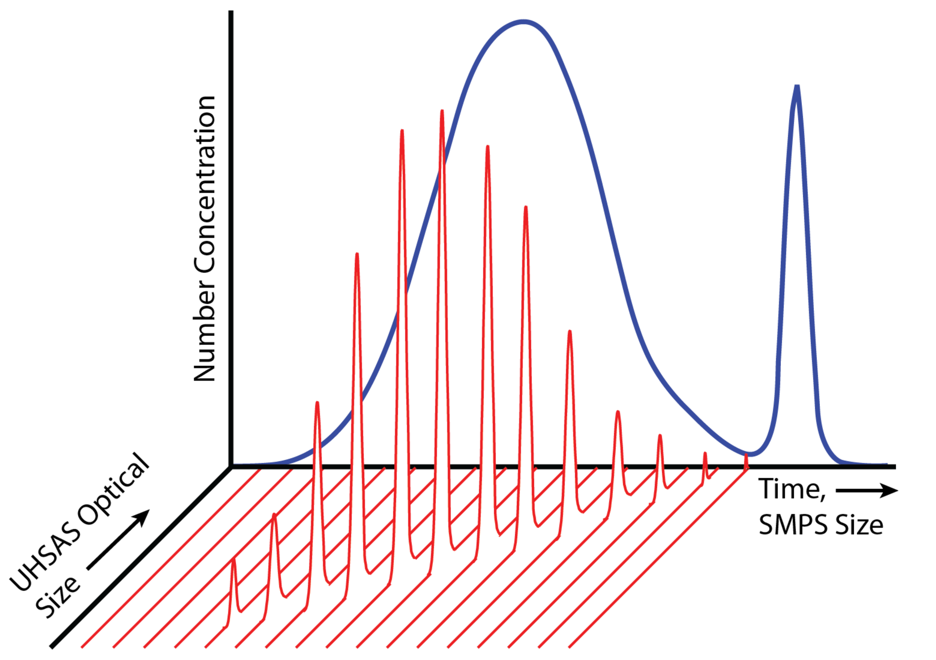


Figure 2. Schematic of the combined optical and mobility size distribution response.

Particles of a known composition are generated by nebulizing ultrapure water (>18 MΩ resistivity). The sample stream is dried in a silica gel diffusion dryer to < 20 % RH as measured by a Vaisala Humicap HMP60 probe. The dry aerosols are size classified with a differential mobility analyzer (DMA; TSI 3081) operated at a sheath-to-aerosol flow ratio of 4.6:1, before being counted by a condensation particle counter (CPC; TSI 3772) and optically sized by one of two optical particle counters (OPCs).

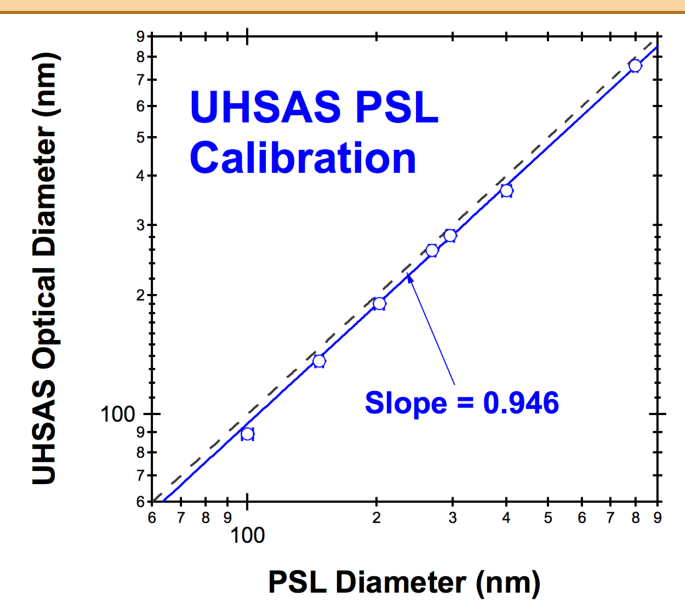
The two investigated OPCs have nearly identical scattering geometries and are as follows:

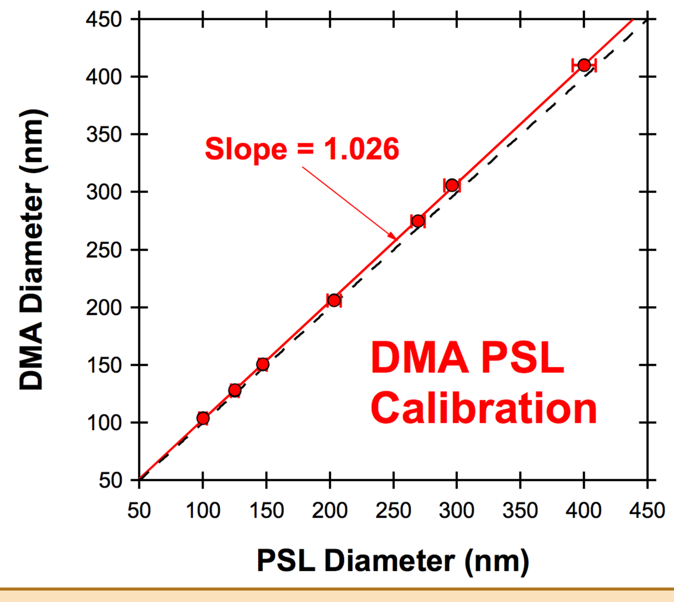
- Droplet Measurement Technologies (DMT) Ultra-High Sensitivity Aerosol Size Spectrometer (UHSAS) measures particles from 0.06 – 1.00 μm diameter. The laser is a Nd^{3+}:YLF solid state laser at 1054 nm with an intra-cavity power of ~1 kW. Scattered light is collected over the angular range of 33-73° and 107-147° (M. Freer, *Personal Communication*, 05 October 2015).
- TSI, Inc. Laser Aerosol Spectrometer (LAS) measures particles from 0.10 – 7.5 μm diameter. The laser is a He:Ne gas laser at 633 nm with an intra-cavity power of ~1-10 W.

Six different organic and inorganic compounds of varying real refractive index and zero imaginary refractive index are investigated and are listed in Figure 5 with their real refractive index values as obtained from the literature. Three additional compounds with non-zero imaginary refractive indices (nigrosine, aquadag, and fullerene soot) are also investigated and are shown in Figure 6. It should be noted that these refractive index values are uncertain due to limited measurements reported in the peer-reviewed literature.

Instrument Calibration

Monodisperse NIST-traceable polystyrene latex spheres (PSL; Thermo Scientific, Inc.) with a real refractive index of 1.59 are used to verify the DMA electrical mobility sizing and to calibrate the UHSAS and LAS optical sizing. All experimental data reported here in the results section are corrected for these minor calibration drifts using the measured slopes in Figure 3.





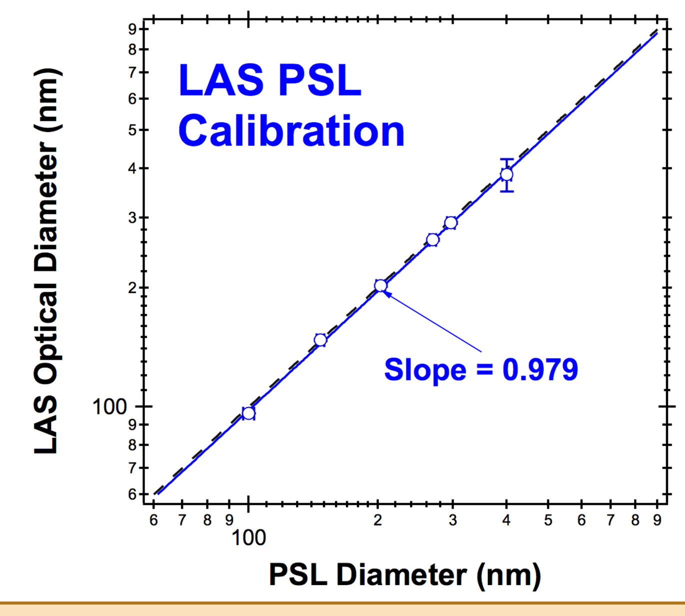


Figure 3. Calibration curves for the DMA, UHSAS, and LAS using mondisperse, NIST-traceable PSLs. Size distribution peaks were fit with Gaussian curves and the data points denote the mean of each peak. Solid, colored lines are linear fits to the data points with the intercept forced through zero. The black, dashed line is the one-to-one line.

Results

- The monodisperse aerosol stream exiting the DMA is sent to the CPC and one of the two OPCs. The DMA size is varied stepwise in time producing distributions like that shown in Figure 5 for ammonium sulfate (m = 1.53) aerosols being measured by the LAS.
- The difference between the DMA mode diameter (white line in Figure 5) and the mode of the LAS size distribution is due to the difference in real refractive index between ammonium sulfate and PSLs.

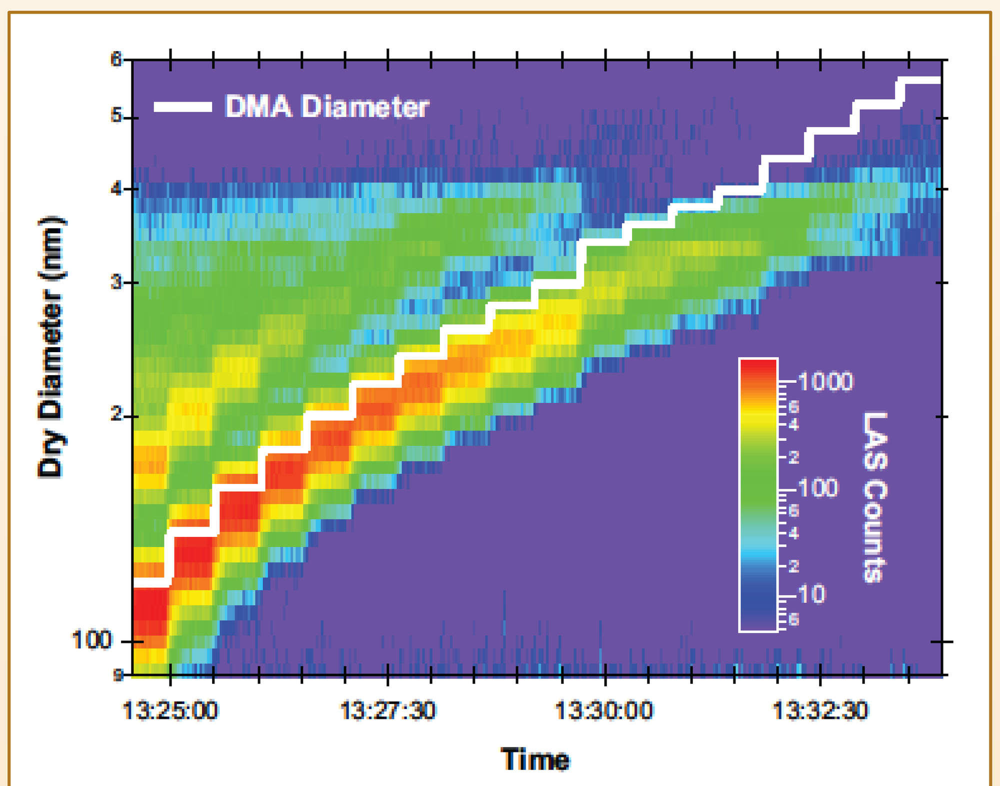


Figure 5. Example LAS size distributions for ammonium sulfate aerosols (m = 1.53) as the DMA voltage (and hence mobility diameter) is increased in a stepwise manner.

- In the top row of Figure 6, mean optical diameter as calibrated to PSL (m = 1.59) is plotted against mean DMA diameter. Consequently, the solid black 1:1 line can be interpreted as a PSL calibration line. Colored lines with points are measurements of non-absorbing organic and inorganic aerosol species, while the three samples shown as only points are absorbing compounds.
- The deviation from the PSL calibration line is caused by differences in the aerosol refractive index that arise from changes in chemical composition.
- The bottom row of Figure 6. shows the percent difference between optical and DMA diameters. Ranges are from ~4-20% for the UHSAS and ~4-28% for the LAS. The three absorbing species are excluded in the percent difference calculation to highlight the variability amongst the non-absorbing compounds and because the nigrosine, aquadag and fullerene soot points are significantly off scale.

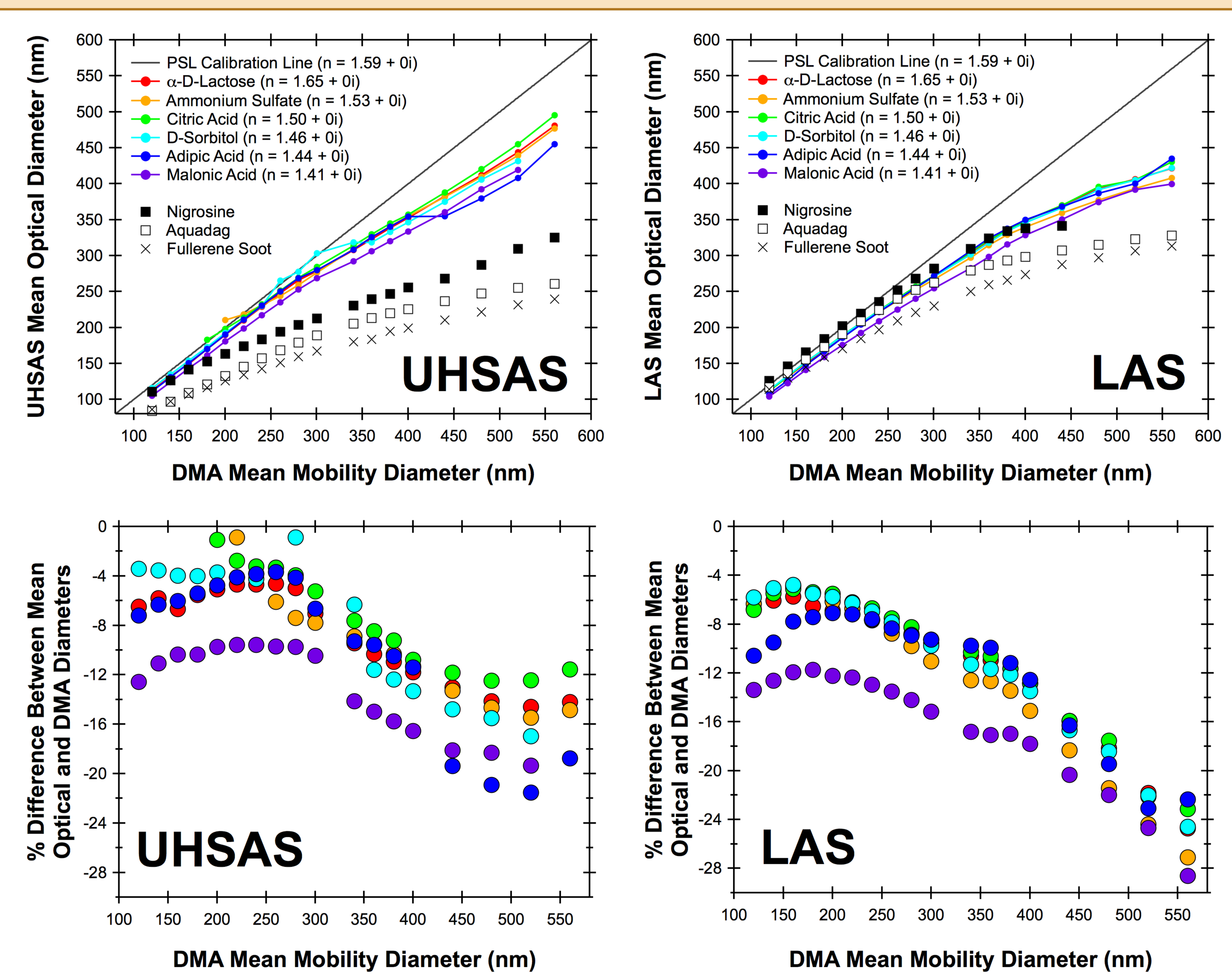


Figure 6. Comparison plots of optical and mobility size for different laboratory-generated aerosol types for the UHSAS (left column) and the LAS (right column). The top row subfigures are 1:1 plots, while the bottom row subfigures show the percent difference in mean size between the optical and mobility sizing techniques for the non-absorbing aerosols. Absorbing aerosol data are omitted from the lower subfigures.

Summary & Conclusions

- In this study, we test the ability of combined electrical mobility and high-resolution optical sizing to distinguish between aerosols of varying refractive index.
- Aerosol compositional changes manifest themselves as a significant under-sizing relative to PSL calibration size standards; however, there does not appear to be sufficient sensitivity of the method for differentiating the individual particle refractive indexes.
- This study shows that users of optical particle counters need to account for the significant changes from decreased real refractive index or increased imaginary refractive index associated with real-world aerosol types. There is also a large sizing bias associated with the UHSAS that is not apparent with the LAS for absorbing aerosol. This may be due to the differences in laser wavelength and intracavity power between the two instruments.

Future Work

We plan to model these empirical results using Mie Theory and explore the use of theory with bulk scattering and absorption measurements to infer particle complex refractive index.

References

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- Hasekamp, O. et al., *J. Geophys. Res.*, 2011
- Mischenko, M. et al., *BAMS*, 2007
- NRC, *Earth Science Applications From Space: National Imperatives for the Next Decade and Beyond*, 2007