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Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
ENA	Eastern North Atlantic
OPC	optical particle counter
PSL	polystyrene latex
UHSAS	ultra-high-sensitivity aerosol spectrometer

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1.0 Introduction

The ultra-high-sensitivity aerosol spectrometer (UHSAS) (Droplet Measurement Technologies, Longmont, Colorado) is an optical particle size spectrometer that determines aerosol size distributions in the diameter range from 60 to 1000 nm in 100 logarithmically spaced diameter bins[1] by measuring the amount of light scattered by individual aerosol particles from a 1054-nm laser. Recently, it was noticed that aerosol size distributions determined by the UHSAS from all U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility sites exhibited modes at diameters greater than ~500 nm (Figure 1, blue arrows). These modes were not present in size distributions determined by another aerosol spectrometer that was co-located with the UHSAS, the Grimm 11D optical particle counter (OPC) (GRIMM Aerosol Technik Ainring GmbH & Co. KG, Germany), which sizes particles into 31 linearly spaced diameters bins from 253 to 35,150 nm using a 683-nm wavelength laser. Additionally, abrupt changes in aerosol particle number concentration with changing particle diameter are seen in UHSAS data at several diameters (Figure 1, red arrows). The purpose of this report is to explain the origin of the modes and the abrupt changes in aerosol size distributions observed by the UHSAS and why they are absent in the data from the Grimm OPC.

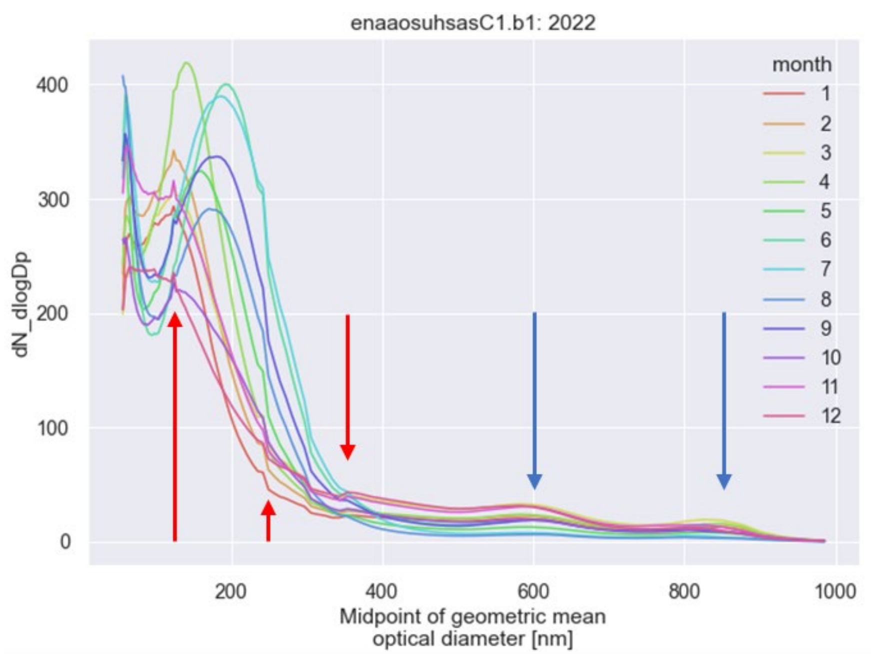

Figure 1. Monthly mean particle size distributions measured by the UHSAS instrument at the ARM Eastern North Atlantic (ENA) site in 2022. Wave-like patterns are visible at ~600 and ~ 820 nm (blue arrows), and abrupt changes in particle number concentration are seen at ~130, ~260, and ~380 nm (red arrows).

[1] The number and distribution of size bins can be selected by the user. The configuration presented here is what ARM uses for their UHSAS instruments.

2.0 UHSAS Calibration

The UHSAS is calibrated using spherical polystyrene latex (PSL) particles with known diameters. The UHSAS internal calibration curve approximates the theoretical (Mie) scattering curve for the amount of scattered light for a given particle diameter and index of refraction[2], which is assumed to be 1.58 for PSLs at 1054 nm. Therefore, the diameter of a spherical aerosol particle with a different index of refraction (typical for ambient aerosol particles) determined by the UHSAS will be different from its geometric diameter[3]. This phenomenon is illustrated in Figure 2, which presents the theoretical response of the UHSAS as a function of particle diameter for PSL particles (with index of refraction 1.58) and for purely scattering spherical particles with indices of refraction 1.4 and 1.5. The response of 2´10^5 (arbitrary units) resulting from a particle with index of refraction 1.5 with diameter near 650 nm (right vertical red line) is the same as the response given by a PSL particle with diameter near 570 nm (left vertical red line), which would be the diameter of the particle reported by the UHSAS. A particle with index of refraction 1.4 and diameter near 830 nm also would provide this same response, and thus would also be classified by the UHSAS as having diameter ~570 nm. This feature is endemic to all optical detectors, and as indices of refraction of ambient aerosol particles are not a priori known, conversion to actual diameters is not possible. Thus, size distributions determined by the UHSAS, and those of any optical detector, are inherently presented in terms of the optical diameter reported by the instrument for particles of given index of refraction – in this case, that of PSL particles.

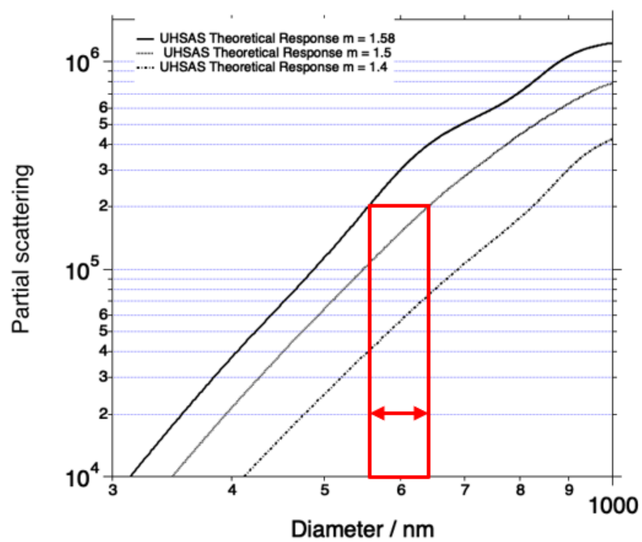

Figure 2. Theoretical Mie scattering curves for different particle indices of refraction, calculated for the UHSAS 1054-nm laser wavelength. The index of refraction of PSL, m =1.58, is used for deriving the UHSAS calibration curve. The red rectangle illustrates the theoretical sizing error from sampling particles with an index of refraction of 1.5 compared to 1.58 used in calibration.

[2] Bohren, CF, and D. Huffman. 2008. *Absorption and scattering of light by small particles.* John Wiley & Sons, New York.
[3] Cai, Y, DC Montague, W Mooiweer-Bryan, and T Deshler. 2008. "Performance characteristics of the ultra-high sensitivity aerosol spectrometer for particles between 55 and 800 nm: Laboratory and field studies." *Journal of Aerosol Science* 39(9): 759–769, https://doi.org/10.1016/j.jaerosci.2008.04.007

3.0 Theoretical Instrument Response

To investigate if the modes present in the UHSAS size distributions result from artifacts of the optical measurement technique, the theoretical aerosol size distribution that would be determined by the UHSAS was calculated for an artificial aerosol size distribution $\frac{\mathrm{d}N}{\mathrm{dlog}D} = \left(\frac{D}{100}\right)^{-3}$ (where D is particle diameter in nanometers). This size distribution was selected because it exhibits no structure (i.e., no modes), but rather a smooth decrease in amplitude with increasing diameter. Therefore, any modes that appear in the size distribution determined (theoretically) by the UHSAS will result from artifacts of the optical measurement process used by the UHSAS. Two different indices of refraction, 1.4 and 1.5, typical of atmospheric aerosols, were investigated. The diameters of the particles determined by the UHSAS were calculated using the theoretical (Mie) scattering curve for aerosol particles and the instrument operating parameters (angular detection regions and wavelength) of the UHSAS, as discussed above with regard to Figure 2. The resulting UHSAS size distributions (Figure 3) exhibit prominent modes near 800 nm (for index of refraction 1.5) and 900 nm (for index of refraction 1.4) similar to those observed in the ambient size distributions from the ARM sites (Figure 1). As the underlying size distribution contained no structure, these modes are artifacts of the optical measurement technique.

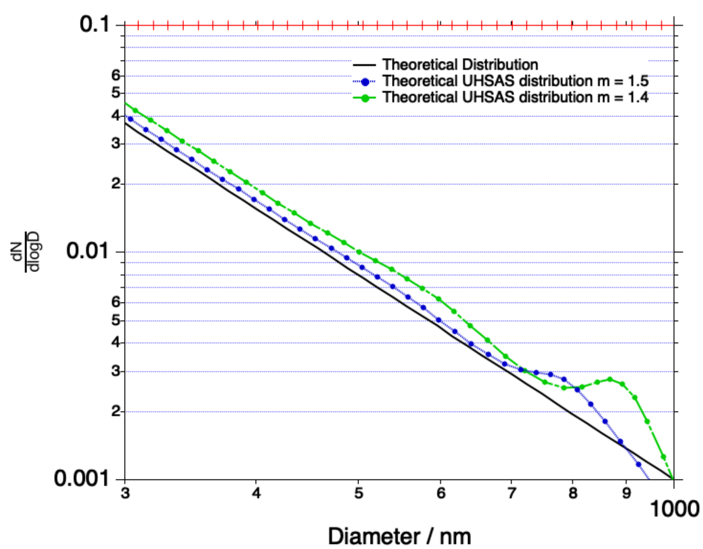

Figure 3. Theoretical ambient size distribution of particles dN/dlogD = (D/100 nm)$^{-3}$ (solid line) and theoretical UHSAS instrument responses (i.e., reported size distributions) for particles with indices of refraction 1.4 and 1.5. The y-axis is on a logarithmic scale to emphasize the modes at higher particle diameters. Red markers on the top axis denote the UHSAS diameter bin limits.

The same theoretical experiment was performed for the Grimm OPC, using its specific operating parameters, in two ways: first by assuming the same diameter bin distribution as that of the UHSAS, and then with the actual (much coarser) diameter bin distribution used by the Grimm OPC. The size distribution that would be reported by the Grimm OPC for the same size bin spacing as that of the UHSAS exhibits several modes for both indices of refraction 1.4 and 1.5, and they occur at lower diameters (Figure 4a) because the wavelength of the Grimm OPC (683 nm) is lower than that of the UHSAS (1054 nm). The size distribution that would be reported by the Grimm OPC using its actual diameter bin spacing (Figure 4b) shows no modes because the sizing resolution is so coarse that any modes are smoothed and essentially missed. This result matches what is seen in the field: the absence of modes in ambient size distributions reported by the Grimm OPC.

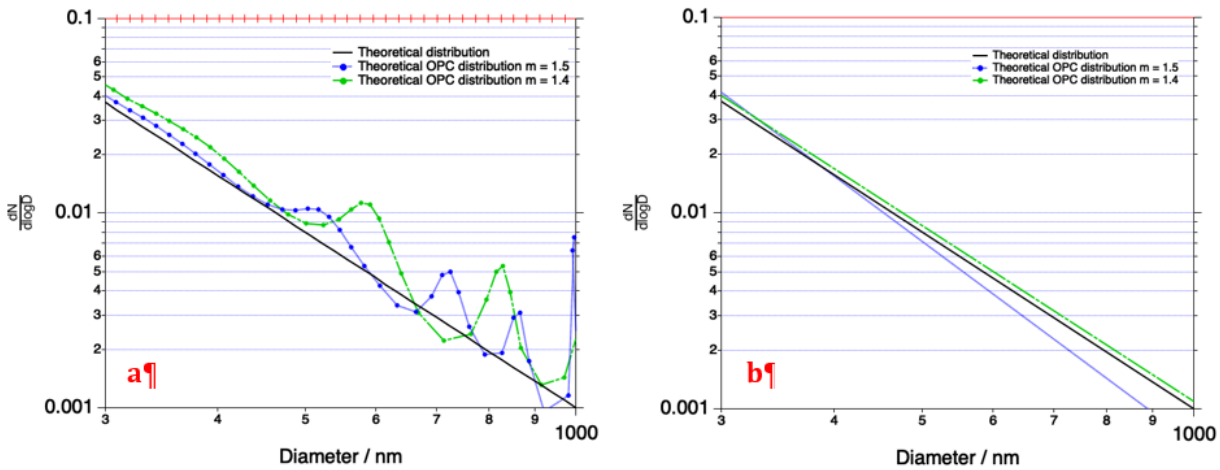

Figure 4. Theoretical ambient size distribution of particles dN/dlogD = $(D/100\ nm)^{-3}$ (solid lines) and theoretical Grimm OPC instrument responses (reported size distributions) for particles with different indices of refraction, using the same size bin distribution as in UHSAS (a) and the actual size bin distribution of the Grimm OPC (b). Red markers on the top axis denote the diameter bin limits for each case. Note that the actual size bin distribution of the Grimm OPC is such that there is only a single diameter bin in the 300-1000-nm range.

The artifacts resulting from the smooth size distribution, when there is sufficiently high sizing resolution, are inherent in the optical detection technique itself, and are not due to measurement error. The location and magnitude of these artifacts will differ among different instruments based on their specific operating parameters, primarily laser wavelength. The apparent modes reported by instruments that use lasers with higher wavelengths will occur at larger diameters than those that use lower wavelengths, as seen by comparing the locations of the modes that would occur in the UHSAS (Figure 3) to those from the Grimm OPC with the same size bin spacing (Figure 4a) for the structureless size distribution selected. The approach of using coarser resolution might eliminate spurious modes from the data, but it does so at the cost of coarser resolution, especially for large sizes where ambient particle concentrations are low but can have large effects on properties such as light scattering and mass concentrations. In addition to differences in operating parameters among instruments, the artifacts will depend on the (unknown) size distributions and properties (i.e., indices of refraction) of the sampled aerosol particles. These effects would be expected to vary between measurement sites and seasons.

4.0 UHSAS Optical Sensor Gain Stages

Additional artifacts in size distributions are reported by the UHSAS, besides those described above, that are inherent to its design and result from the use of multiple light sensors with different sensitives. Multiple sensors are required because the amount of light scattered by a particle depends strongly on its diameter. For instance, at wavelength 500 nm the scattering cross-section of a particle with index of refraction 1.5 at wavelength 500 nm varies by more than five orders of magnitude as the diameter increases from 60 nm to 1000 nm (the measurement range of the UHSAS). As this is much greater than the dynamic range of a single optical sensor, optical instruments typically use multiple sensors and multiple "gain stages" to overcome this issue. The UHSAS uses two physical sensors with different light sensitivities, each of which has two amplification levels for a total of four gain stages. Each of these gain stages covers only part of the range of scattered light intensities and thus range of diameters reported by the UHSAS. As it is not possible to adjust the individual sensors to perfectly align the measurements of adjacent gain stages, the overall size distribution is obtained by "stitching" together the measurements from individual gain stages,[4] which may result in abrupt changes in particle number concentrations between adjacent diameter bins belonging to different gain stages. The stitching points between adjacent gain stages are specific to individual UHSAS units and thus the corresponding features seen in the UHSAS data will vary between instruments. As with the apparent modes described above, these artifacts are unavoidable and are intrinsic to the UHSAS operation.

5.0 Summary

Aerosol size distributions determined by optical techniques contain artifacts that are inherent in optical measurements. The apparent modes seen in the UHSAS aerosol size distribution data collected at various locations also occur for size distributions with no structure and are absent in size distributions reported by other optical detection instruments only because these instruments have coarse sizing resolution. The abrupt changes in the UHSAS size distributions at different diameters are also inherent in optical detection techniques due to the large range of the signal detected by the instrument over a wide diameter range, resulting in the necessity of multiple gain stages. These abrupt changes can be minimized by adjusting the measurements of overlapping gain stages, but they cannot be entirely removed. Care should be taken when interpreting size distributions determined from any optical instruments so that the artifacts from the instrument operation are not attributed to properties of the ambient aerosol.

[4] Moore, RH, EB Wiggins, AT Ahern, S Zimmerman, L Montgomery, PC Jost, CE Robinson, LD Ziemba, EL Winstead, BE Anderson, CA Brock, MD Brown, G Chen, EC Crosbie, H Guo, JL Jimenez, CE Jordan, M Lyu, BA Nault, NE Rothfuss, KJ Sanchez, M Schueneman, TJ Shingler, MA Shook, KL Thornhill, NL Wagner, and J Wang. 2021. "Sizing response of the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) and Laser Aerosol Spectrometer (LAS) to changes in submicron aerosol composition and refractive index." *Atmospheric Measurement Techniques* 14(6): 4517–4542, https://doi.org/10.5194/amt-14-4517-2021

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