Supplement of

Sizing Response of the Ultra-High Sensitivity Aerosol Size Spectrometer (UHSAS) and Laser Aerosol Spectrometer (LAS) to Changes in Submicron Aerosol Composition and Refractive Index

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Description of Supplementary Data Included in Moore_AMT_LAS_UHSAS_SupplementaryData_v1.h5:

The data points shown in Figures 5-7 of the main text are included as one-dimensional data objects within the HDF5 file in this supplement. HDF5 (https://www.hdfgroup.org/) is a hierarchical data format, which can be opened using the HDFView software (https://www.hdfgroup.org/downloads/hdfview/) as well as commonly-used data analysis programs such as MathWorks Matlab (https://www.mathworks.com/help/matlab/hdf5-files.html) and WaveMetrics Igor Pro (https://www.wavemetrics.com/products/igorpro/dataaccess/hdf5). The data object format for Figures 5 and 6 is of the form, <**Compound**>_<**MMDD**>_<**Instrument**>_**Dp**, where **Dp** denotes the mean diameter and

<**Compound**> is the abbreviated chemical species: ammonium sulphate (AS), fullerene soot (Fullerene), lithium bromide (LiBr), lithium fluoride (LiF), sodium chloride (NaCl), sodium fluoride (NaF), nigrosine dye (nigrosine), Pahokee Peat Humic Acid (PPHA), polystyrene latex spheres (PSL), Suwannee River Fulvic Acid (SRFA), and Suwannee River Humic Acid (SRHA)

<MMDD> is the two-digit month and two-digit date of the data collection in 2020

<Instrument> denotes the differential mobility analyser (SEMS) mobility diameter set point or the mean peak diameters for the LAS or UHSAS. The instrument is omitted for the manufacturer-specified PSL mean diameters.

For the Figure 7 data objects, the format is of the form,

<ProjectYear>_<Compound>_<MMDD>_<Instrument>_<DiameterType>, where

<**ProjectYear**> denotes either the DC-3 field data collecting 2012 (PY2012) or the lab data collected in 2020 (PY2020)

<**Compound**> is the abbreviated chemical species: ammonium sulphate (AS) or polystyrene latex spheres (PSL)

<MMDD> is the two-digit month and two-digit date of the data collection in 2020; omitted for 2012 data

<Instrument> denotes the differential mobility analyser (SEMS) mobility diameter set point or the mean peak diameters for the LAS or UHSAS. The instrument is omitted for the manufacturer-specified PSL diameters.

<DiameterType> denotes the mean diameter (Dp and Dp_avg) or the diameter standard deviation reported by the PSL manufacturer (Dp_stdev)

Supplementary Mie Theory Figures:

Additional Mie theory calculations are performed to compare the theoretical instrument response calculated both using the UHSAS optical collection angles and for the integrated light scattering across all angles. As can be seen from Figures S1-S4, the assumed optical collection geometry becomes more important for larger particles approaching and exceeding one micron in diameter. While performing the optical calculations using the actual optical geometry is preferred, this analysis shows that simplified calculations using integrated scattering quantities reasonably approximate the instrument performance for atmospherically-relevant, accumulation mode aerosols.

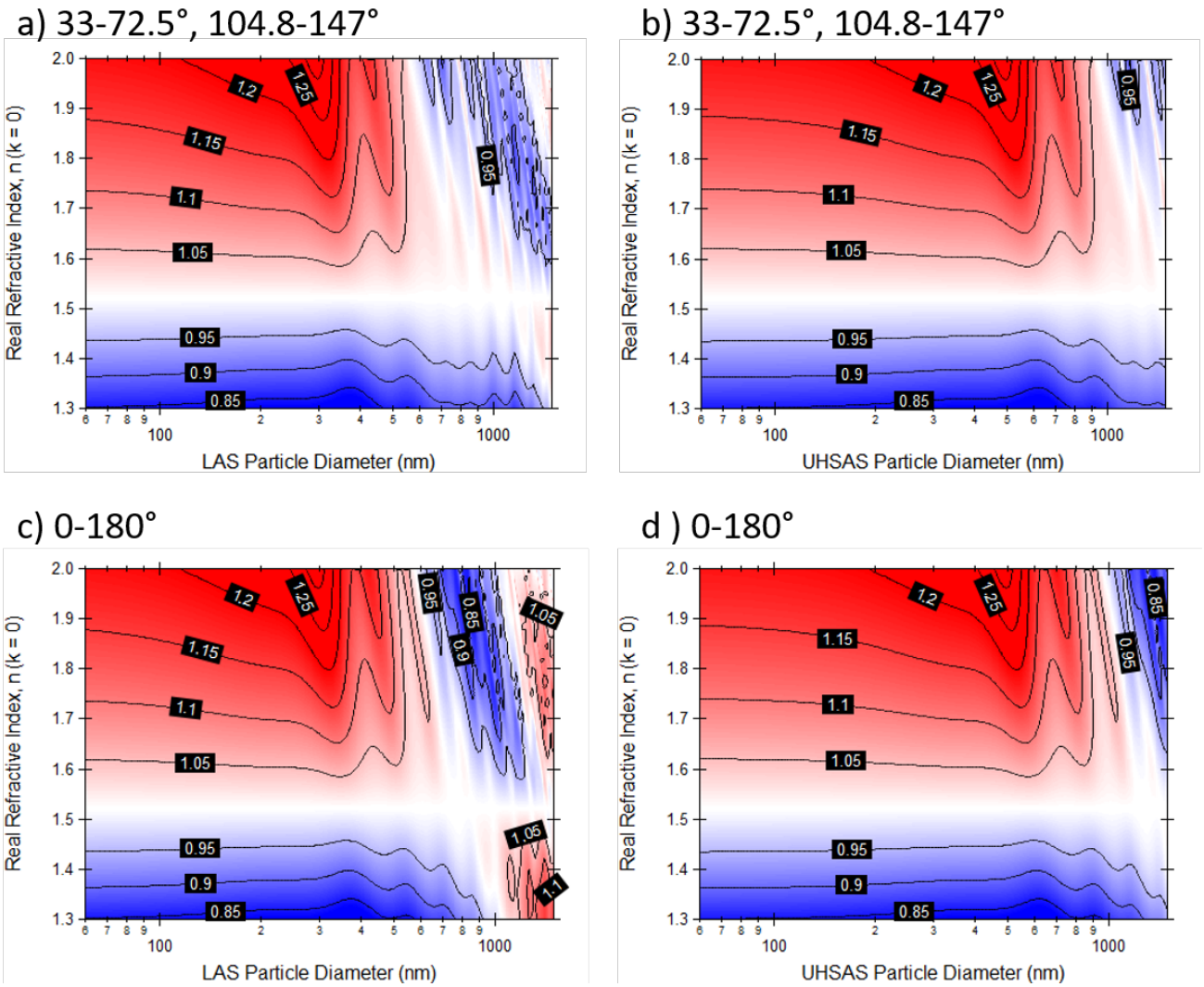

Figure S1: Mie theory calculations of size-resolved particle scattering normalized to that for ammonium sulphate (1.52+0i) at the LAS laser wavelength (a, c) and the UHSAS laser wavelength (b, d), where the imaginary part of the refractive index is held constant at zero. Panels a and b are those shown in Figure 1 of the main text using the UHSAS collection optics geometry (33-72.5° and 104.8-147°), while panels c and d are calculated for the total integrated scattering across the entire phase function (0-180°). The coloured shading represents the ratio of the theoretical particle scattering cross section (on the same colour scale as in Figure S2), while the contours approximate the expected sizing ratio, which is assumed to be equal to the sixth root of the scattering cross section ratio (reasonable for diameters below 300-600 nm).

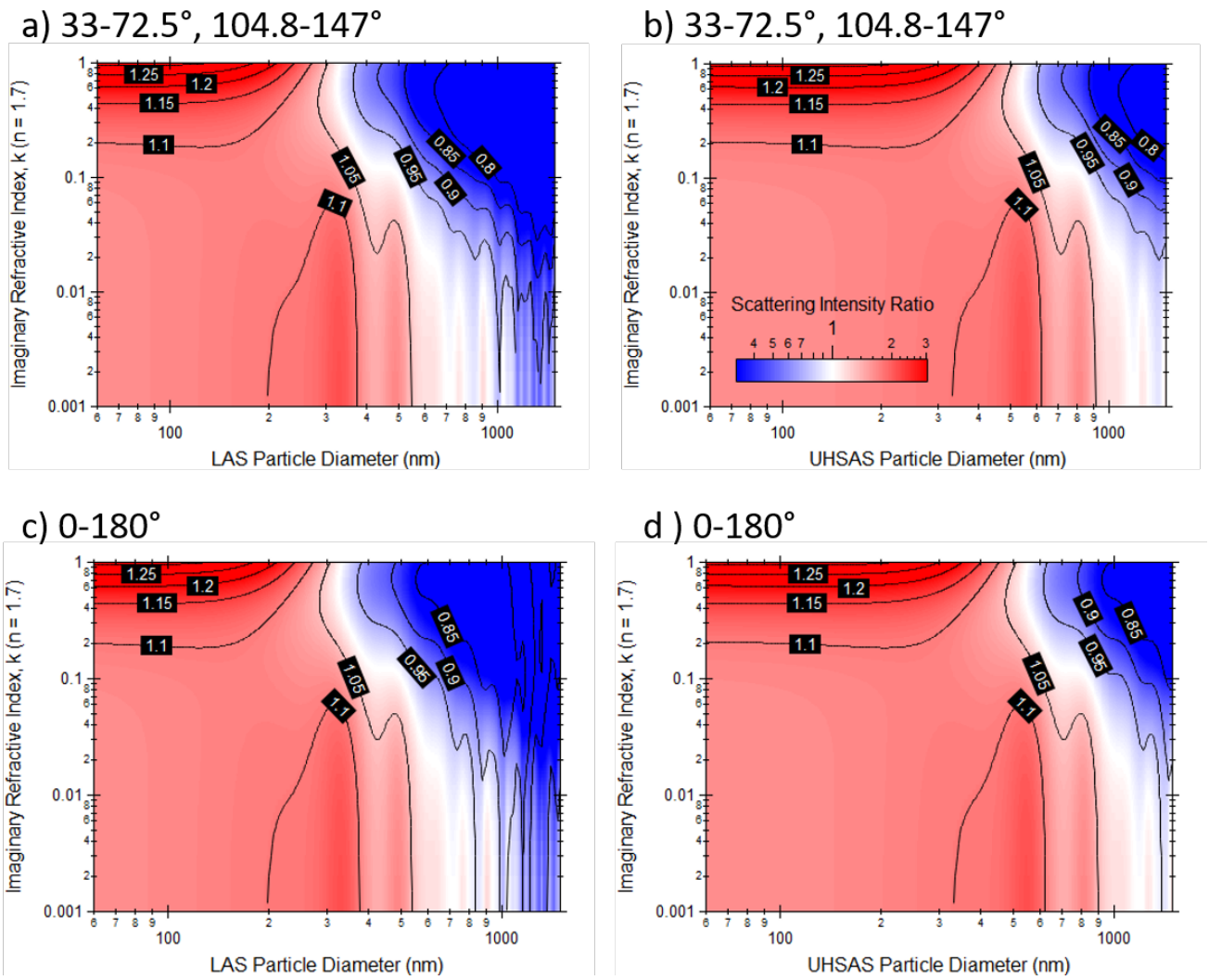

Figure S2: Mie theory calculations of size-resolved particle scattering normalized to that for ammonium sulphate (1.52+0i) at the LAS laser wavelength (a, c) and the UHSAS laser wavelength (b, d), where the real part of the refractive is held constant at 1.7. Panels a and b are those shown in Figure 1 of the main text using the UHSAS collection optics geometry (33-72.5° and 104.8-147°), while panels c and d are calculated for the total integrated scattering across the entire phase function (0-180°). The coloured shading represents the ratio of the theoretical particle scattering cross section, while the contours approximate the expected sizing ratio, which is assumed to be equal to the sixth root of the scattering cross section ratio (reasonable for diameters below 300-600 nm).

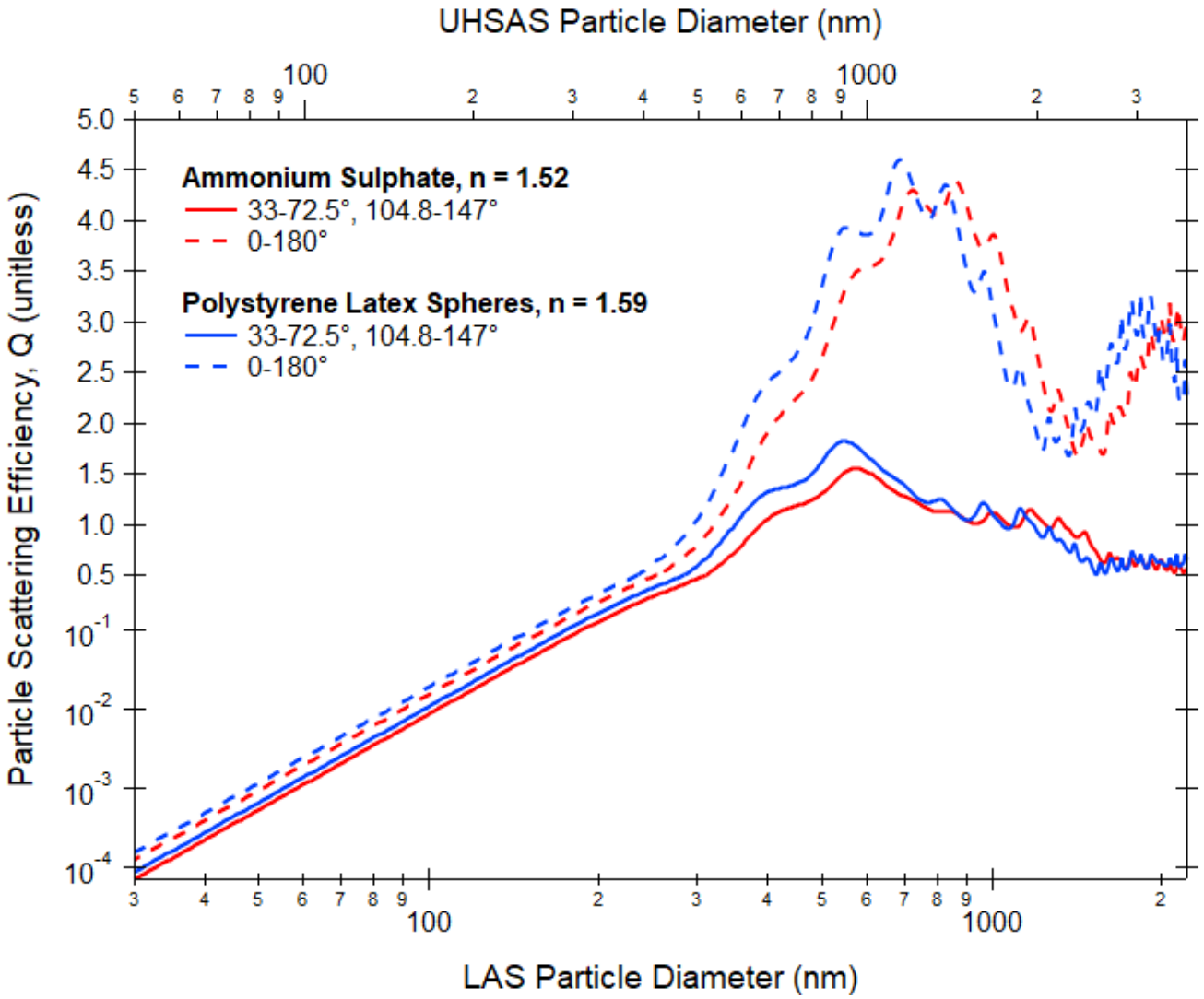

Figure S3: Mie theory calculations of size-resolved particle scattering efficiency for purely scattering particles with refractive indices the same as ammonium sulphate (1.52+0i) and polystyrene latex spheres (PSLs; 1.59+0i). Solid curves are calculations using the UHSAS collection optics geometry (33-72.5° and 104.8-147°), and dashed curves are calculations for the total integrated scattering across the entire phase function (0-180°).

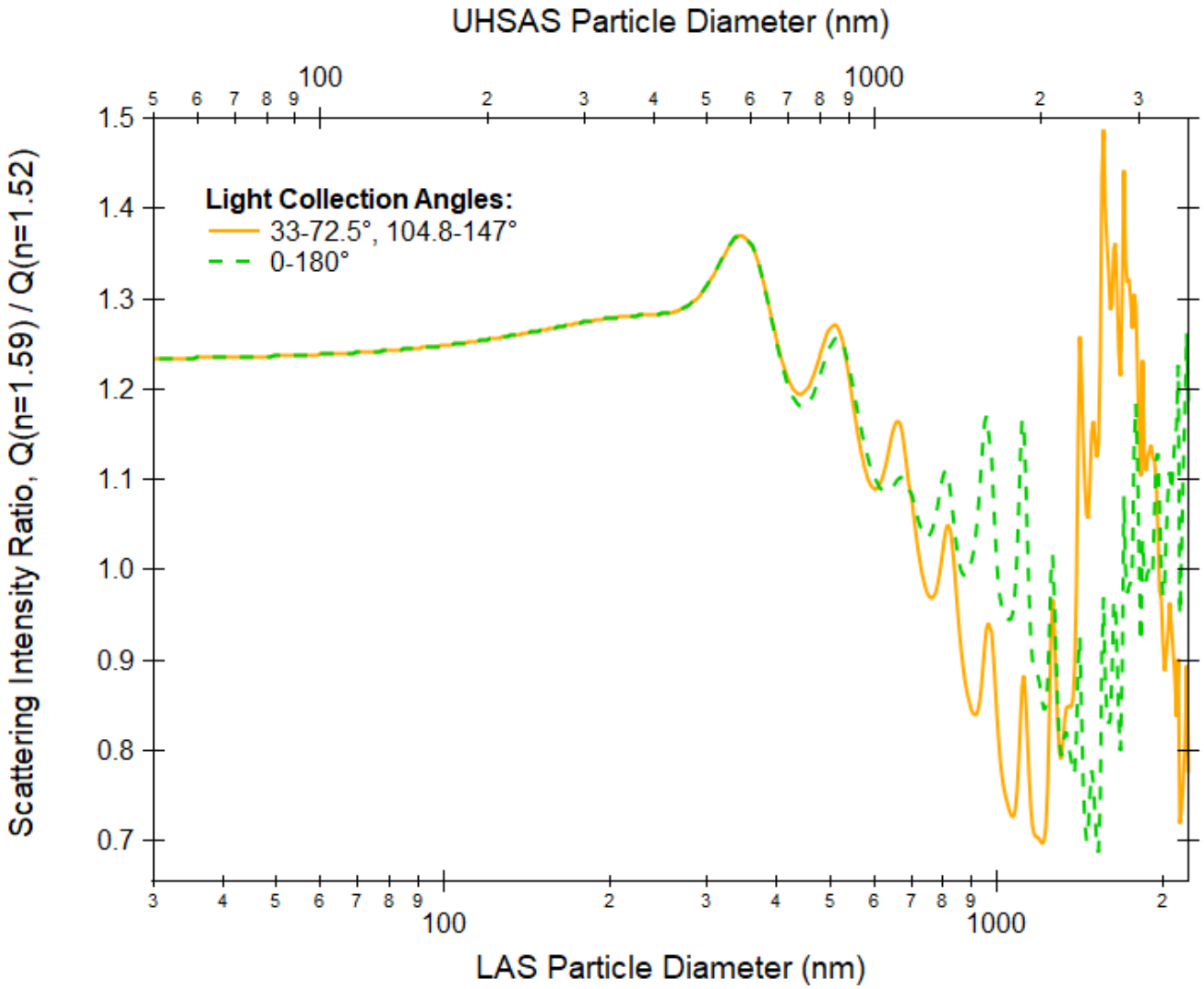

Figure S3: Mie theory calculations of size-resolved particle scattering for polystyrene latex spheres (PSLs; 1.59+0i) normalized to that for ammonium sulphate (1.52+0i) for the UHSAS laser wavelength (top axis) and LAS laser wavelength (bottom axis). The solid, orange curve is calculated using the UHSAS collection optics geometry (33-72.5° and 104.8-147°), while the dashed, green curve is calculated for the total integrated scattering across the entire phase function (0-180°).