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Reply to Interactive comment on "Determination of the refractive index of insoluble organic extracts from atmospheric aerosol over the visible wavelength range using optical tweezers" *by*

Rosalie H. Shepherd et al.

Anonymous Referee #1

Interesting experimental work dealing with the characterization of aerosol particles where the imaginary part of the refractive index cannot be neglected.

Thank you

Questions/comments that should be addressed:

1. To echo what the other two reviewers stated: It seems like some important references regarding the determination of the refractive index of single aerosol particles using Mie theory are missing from the introduction.

The following references have been added:-

R.E.H. Miles, J.S. Walker, D.R. Burnham and J.P. Reid, 'Retrieval of the Complex Refractive Index of Aerosol Droplets from Optical Tweezers Measurements', Physical Chemistry Chemical Physics 14 (2012) 3037–3047.

H.-B. Lin, J.D. Eversole, A.J. Campillo, "Identification of morphology dependent resonances in stimulated Raman scattering from microdroplets", Optics Communications, 77(5,6) (1990) 407-410

With the following text added to the paper:

"The use of morphological dependent resonances in Raman Spectra to determine refractive index at a fixed wavelength has been reported by Lin et al. (1990) and references therein and Miles et al. (2012)."

2. Were the Mie spectra calculated by integrating over the acceptance angle of the objective? Please add a sentence or two that qualitatively describes the calculation. On page 3 it is stated "simulating the spectrum with Mie calculations." That is too vague.

Yes, the following text has been changed to read:-

"The optically trapped aerosol droplet was illuminated with white light and the elastically backscattered light was collected over a 25-degree cone angle, by an

objective lens. Further optics (described in Jones et al, 2013) focused the light onto a spectrometer (Acton, SP1500i)."

and

"The measured Mie spectrum was simulated through the application of Bohren and Huffman(1983) formalism of Mie theory integrating over 25 degree cone angle of backscattered light. In conjunction with the Cauchy Equation..."

3. Can you provide more details on how the size and Cauchy parameters were found. A simple grid search? If so, over what space? Was the imaginary part of the refractive index included in the search space?

The following text has been added:-

"Typically, the radius of droplet was fixed and the values of A, B, and C varied until a good fit between measured and simulated Mie spectra was achieved by simple comparison (inspection) of peak, trough and inflection point positions. The value of the radius was then iterated through a series of radii with optimization of the values of A, B. and C as a function of wavelength repeated at each radius. Thus, a qualitative grid search was performed over parameter space. Parameter space was A varying from 1.3 to 1.7, B from 0 to 20,000 nm⁻² and C from 0 to 1×10^9 nm⁴. The value of the radius will be between 0 to 3 microns typically. The imaginary component of the refractive index was varied only after the grid search for the woodsmoke and humic acid samples."

The Imaginary part of the refractive index was described by an Ångström equation. The value of the Ångström exponent was calculated from the UV-Vis Absorbance measurement of the bulk sample and shown to be consistent with the attenuated Mie Spectra in figure 2. The issue and changes to text are further addressed in our answer to point 6, from reviewer 3.

4. In Section 4.4, the uncertainty associated with fitting less structured Mie spectra is mentioned. What is the origin of this increased uncertainty when fitting Mie spectra of absorbing particles or submicron particles studied at optical wavelengths? Presumably, Mie theory allows you to calculate the observed spectrum very accurately. Yet, when sharp peaks are absent, the uncertainty in the retrieved parameters increases substantially. A thorough answer to this question is not necessary but a slightly more detailed comment in the text would be helpful.

The following text has been added:-

"The Mie spectra in figure 1 of Summer Urban aerosol extract are structured with pronounced peak shapes that allow the facile fitting between simulated and measured Mie Spectra. Such spectra allow a relatively small range of values of radius, Cauchy coefficients A, B and C to provide a good fit between measured and simulated Mie Spectra. The rest of the Mie spectra in Figure 1 have significantly fewer Mie resonances and their peak shapes are less pronounced. The uncertainties become larger as the spectra become less structured."