

Reply to comments of Reviewer #3:

We would like to thank the Reviewer for his/her helpful remarks. Below, please find our detailed point by point replies to the comments made by the Reviewer.

- 1) The fitting procedure to get the absorption coefficient, the SSA and the imaginary part of the RI depends on one direct absorption measurement, the PAS measurement at 405 nm which has a large error (stated to be typically 60 %). So, to get the values for these properties over the full wavelength range, the authors must assume a variety of power laws or exponential relationships with wavelength to relate the extinction, scattering and absorption measurements. This is very briefly described in section 2.3.1. This is a key part of the retrieval and I think more discussion and consideration of errors needs to be included here, a figure needs to be shown including the trial power laws etc. and the variation in the fitting error. This extra detail would be helpful to the reader.

The PAS measurement error at 404 nm was not stated at 60% error (this was addressed in response to comment 9 of reviewer #1). In fact, Lack et al., (2012) stated that the instruments uncertainty is probably lower than 5%. We acknowledge the fact that the discussion of uncertainty analysis was not clear enough. To address this issue, sections 2.3.1 and 2.3.2 were revised and the discussions is now more elaborated and includes more details relating to the uncertainty propagation of the retrieval.

- 2) Although their retrievals broadly match their expectations for the trends (e.g. in figures 6 and 7), the impact of the errors on the SSA, single wavelength absorption measurements, extinction and scattering measurements on the retrievals need to be more fully discussed. For example, on these two figures, I would expect to see error envelopes on the lines shown in each of these plots, following from the cumulative effect of the uncertainties in the individual measurements.

To address the issue of uncertainty propagation in the retrieval procedure, sections 2.3.1 and 2.3.2 were revised and they now include more details . The calculated uncertainty envelops were added to figures 6a and 7a (now 5a and 6a).

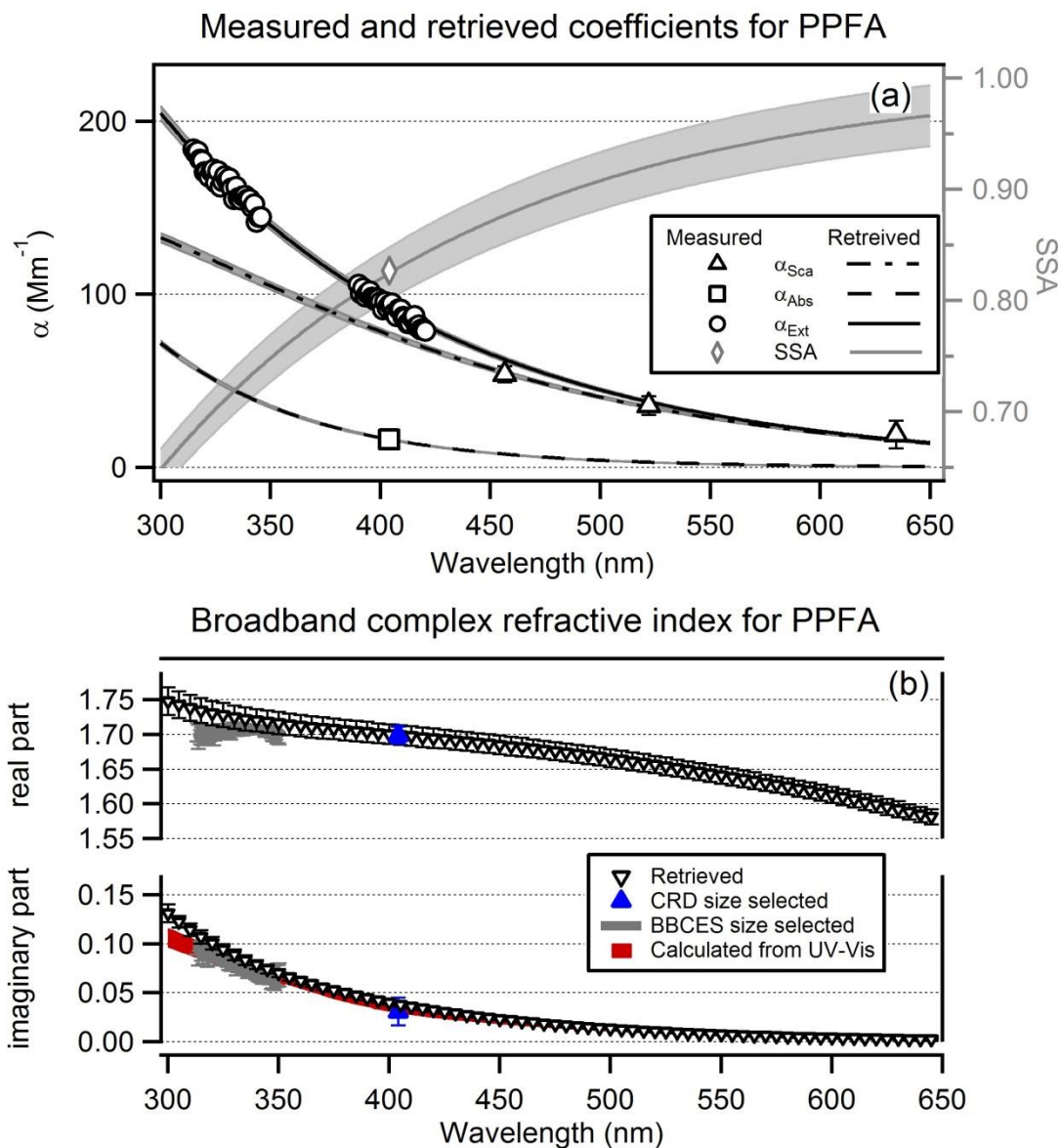
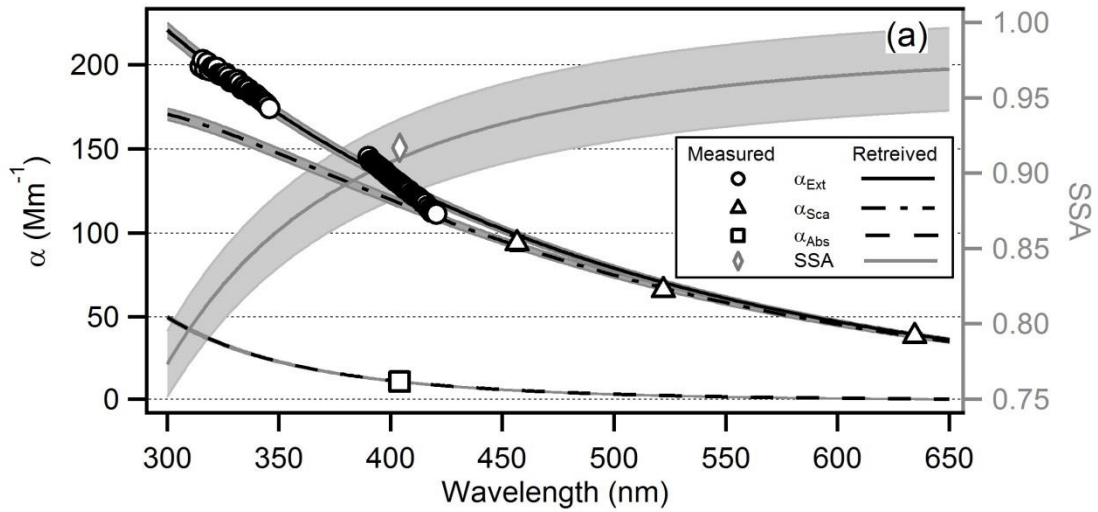


Figure 5. (a) Measured extinction (circles), scattering (triangles), and absorption (square) coefficients (α_{ext} , α_{sca} , and α_{abs} , respectively) for Pahokee peat fulvic acid (error bars represent measurement standard error are partially smaller than the symbols). The retrieved values for broadband extinction (black line), scattering (dash-dot line), absorption (dashed line) and single scattering albedo (SSA; grey line) are also shown with shaded areas represent propagated uncertainty. (b) Retrieved broadband complex refractive index for Pahokee peat fulvic acid using: 1) the retrieved RI from the data shown in panel (a) (inverted triangles); and 2) size selection measurements for the broadband cavity-enhanced spectrometer (BBCES-315; grey line) and the cavity ring down spectrometer (CRD-S) at 404 nm (blue triangles). The imaginary part of the refractive index calculated from UV-Vis absorption measurements is indicated by the red shaded area.

Measured and retrieved broadband coefficients for SRFA



Broadband complex refractive index

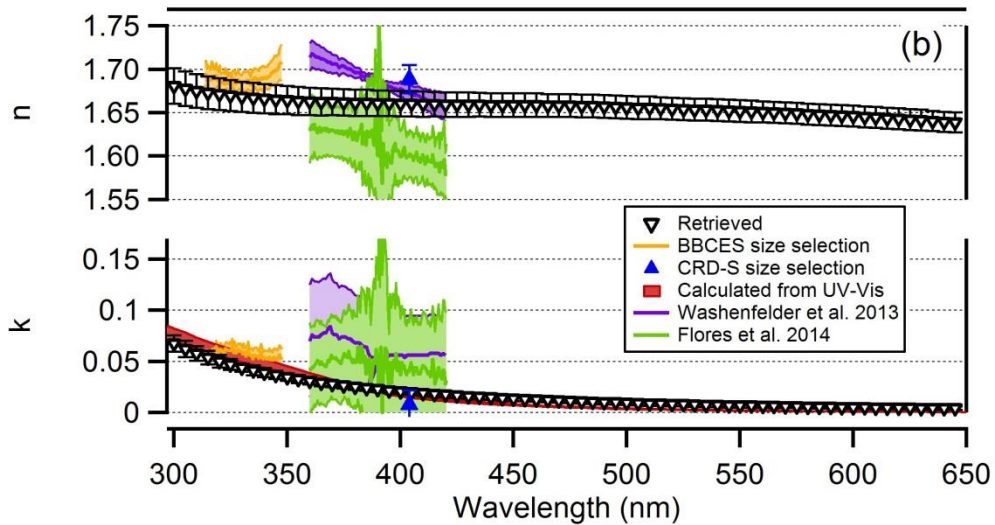


Figure 6. (a) Measured extinction (circles), scattering (triangles) and absorption (square) coefficients (α_{ext} , α_{sca} , and α_{abs} , respectively) for Suwannee river fulvic acid (error bars representing measurement standard error are smaller than the symbols). The retrieved broadband extinction (black line), scattering (dash-dot line), absorption (dashed line) and single scattering albedo (SSA; grey line) are also shown with shaded areas represent propagated uncertainty. (b) Retrieved broadband complex refractive index (RI) for Suwannee river fulvic acid using: 1) the retrieved RI from the data shown in panel (a) (inverted triangles); 2) size selection measurements for the broadband cavity-enhanced spectrometer (BBCES-315; orange line) and the cavity ring down spectrometer (CRD-S) at 404nm (blue triangles); and 3) from the published data of Washenfelder et al. 2013 (purple line) and Flores et al. 2014 (green line). The imaginary part of the refractive index calculated from UV-Vis absorption measurements is indicated by the red shaded area.

Even with the errors reported in section 3.1 from the simulations (of order 5, 10 and 60 %), I would expect the error envelopes to become large when outside the range of the measurements, for example in the retrieved value of the absorption coefficient below 400 nm in Figs. 6 and 7, particularly when the uncertainties in the nephelometer and PAS measurements are considered.

As for the possibility of increased uncertainty outside the range of measurement, we should mention that figures 6a and 7a (now 5a and 6a) show the retrieved optical coefficients which are the fitted curves. As such the uncertainty on each individual point (wavelength) depends on the uncertainty of the fitted coefficients and not on the distance from the measured data point.

These error envelopes should then be included in plots such as Figure 7(b) for the imaginary part of the refractive index. In this plot, error bars are shown in the real part but not the imaginary part, is there a reason for this?

Uncertainties are also included in figures 6b and 7b (the retrieved complex RI) (now 5b and 6b) as error bars, there were simply too small to see with the size of the symbol. The two figures were changed to show the error bars more clearly. In Fig 7 there is a systematic error in the agreement between the retrieval of the extinction coefficient and the measured value around 400 nm with the BBCES data crossing the retrieved line– does this have consequences for the fit? To reduce errors, ideally there should be more overlap in wavelength between the measurements of scattering coefficient and extinction coefficient, what prospect is there for doing this?

We completely agree with the reviewer's perspective on this matter. This could also allow for direct absorption calculation in wavelengths outside the current PAS measurement wavelength. Unfortunately current available nephelometers provide measurements at three wavelengths at the most and our CES instruments were dedicated to measure extinction at lower wavelengths where brown carbon light absorption may be significant. However, we show here conceptually how such measurements can be done and with availability of instrumentation, this method can be implemented to deduce retrievals with lower errors.

Depending on the errors on the imaginary part in Figs 6 and 7, can the authors now definitely state that their new measurements of the optical constants should be preferred over the previous measurements.

Previous studies measured only extinction and it has been shown that adding additional direct measurements of relevant properties such as absorption or scattering improves the accuracy of the retrieval (Zarzana et al., 2014).

Additionally, there is an agreement between our broad band retrieval and the UV-Vis retrieval.

- 3) I think the authors should stress in the abstract that continuous measurements of extinction coefficients are made between 315 and 345 nm and 390 to 420 nm, but

other measurements are pointwise (404, 457, 525, 637 nm). The authors suggest that their approach gives optical coefficients and SSA over the full range 300 – 650 nm but this is not the measurement range. There does seem to be some confusion in the text over the upper wavelength range for the BBCES measurements: the upper range stated in 2.1 (360 to 390 nm) does not match with the range in 2.2.1 (390 to 420 nm). Also line 9 of page 11 says 360 to 390 nm. The abstract was revised and it now addresses the reviewer’s concerns. Additionally, some typos regarding the BBCES measurement range were corrected throughout the manuscript.

- 4) Line 7, page 7: for clarity, specify what the “individual wavelengths” are. This line was revised to: “the effective complex RI of the total particle size distribution is retrieved at each individual wavelength (300 to 650 nm)”.
- 5) Line 27, page 7: The real part of refractive index ranges from 1.692 at 300 nm (presumably this is the wavelength?) and 1.856 at 650 nm. These are much larger values than those expected for SOA, why have such large values been chosen? Why are values for imaginary parts of refractive index quoted to nearest 10^{-6} in magnitude – presumably the measurement is not this sensitive? We thank the Reviewer for this point. The value of 1.856 is a typo, and it is now revised to 1.586. The real part values chosen for the simulation are within current knowledge (Moise et al., 2015). In this manuscript, imaginary parts of the refractive indices are quoted in a magnitude of 10^{-3} , not 10^{-6} .
- 6) Line 3, page 8: An additional error with normal distribution of +/- 2% standard deviation was added to simulated data set. Why this level of error? To address the Reviewer’s concern, the following sentence was added: “This value of error represents typical uncertainty values associated with our instrumental measurements.”
- 7) On page 10 line 20, the authors state “the retrieval of the effective complex RI is strongly dependent on accurate representation of the size distribution and aerosol particle number concentration.” As a consequence they have only used data taken during the night when variations in number concentration are less severe. For the instrument to be robust in the field, is there a solution to this? This is a good point. This can be done by adding a large residence time volume to damp aerosol concentration variation to correspond to the measurement time of the slowest instrument, in this case the SMPS scan time. The following sentence was added: “In field applications, a large volume with long residence time of the sampled air can be added to the system to reduce variations in aerosol concentration.”

Lack, D. A., Richardson, M. S., Law, D., Langridge, J. M., Cappa, C. D., McLaughlin, R. J., and Murphy, D. M.: Aircraft instrument for comprehensive characterization of aerosol optical properties, part 2: Black and brown carbon absorption and absorption enhancement measured with photo acoustic spectroscopy, *Aerosol Sci Tech*, 46, 555-568, 2012.

Moise, T., Flores, J. M., and Rudich, Y.: Optical properties of secondary organic aerosols and their changes by chemical processes, *Chemical reviews*, 115, 4400-4439, 2015.

Zarzana, K. J., Cappa, C. D., and Tolbert, M. A.: Sensitivity of aerosol refractive index retrievals using optical spectroscopy, *Aerosol Sci Tech*, 48, 1133-1144, 2014.