

Reply to comments of Reviewer #1:

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) What are the detection limits for the photoacoustic spectrometer and the cavity ring down spectrometer?

Our cavity ring down aerosol spectrometer (CRD-S) has the following properties: the length of the cavity between the mirrors is 0.95 m, the ratio between the cavity length and the length fill with aerosols (L/d) is 1.193. The typical empty cavity decay time (τ_0) is 30 usec, exponential fitting residual at $t=5*\tau_0$ is less than $\pm 10\%$ (typically $\pm 5\%$), τ_0 standard deviation (std) is about 0.02% (± 0.006 usec). The limit of detection (LOD) defined as $LOD = \tau_0 + 3*Std$ calculates to be 0.055 Mm^{-1} , the limit of quantification (LOQ) defined as $LOQ = \tau_0 + 10*Std$ is 0.185 Mm^{-1} . The photoacoustic spectrometer base line is about 7.5 mV with std of 4.5 mV. Taking the average of 120 measurements at 1 Hz sampling rate (2 minute long measurement) the standard error is 0.411 mV. With calibration slope of $48 \text{ V}^{-1} \text{ Mm}^{-1}$ the LOD is about 0.06 Mm^{-1} and LOQ is about 0.2 Mm^{-1} .

- 2) There probably is not a lot of difference between using an exponential function and using a power law function to extrapolate absorption, scattering, and extinction (the authors can correct me if I am wrong). However, the power law function is more commonly, almost exclusively, used for all three measurements. Can the authors elaborate some on why they choose to employ combinations of both functions?

For the same raw data, power law fitting would probably be slightly steeper than exponential decay function. It is correct that when fitting the same data, the difference between the two methods is not large, but often one approach provides a better fit than the other. From Mie theory calculation, power law dependency of the extinction and absorption with spectrum is expected for small particles (up to 100-300 nm diameters). For larger particles (and/or shorter wavelengths), the ripple structure of the Mie curve is expected to decrease the power law behavior of the spectral extinction and absorption curve. The exponential function is only intended to allow for increased quality of fitting.

- 3) Related to point #2, assuming either an exponential or a power law function holds over the entire spectral range could introduce errors. For example, Massabo et al. [1] recently found that ambient aerosol absorption is better fit to a two- or three-power law function. Given that the current work extrapolates absorption from a single wavelength (404 nm), the choice of extrapolation function could have a sizable impact on the fitting and refractive index retrieval. It is worth noting that such an impact would not be obvious in the PPFA and SRFA samples since they have negligible black carbon components. Consequently, the error could impact

the ambient measurements, though there are no independent measurements that could identify or constrain this potential error.

Technically in this study we extrapolate the absorption from 4 wavelengths and not from one. The absorption measurement at 404 nm is used together with the scattering measurements at additional 3 wavelengths. The extrapolation technique and the error analysis are thoroughly discussed in sections 2.3.1 and 2.3.2.

Additionally, black carbon is not expected to significantly increase the errors using the extrapolation approach since it is also treated with a power law behavior of the absorption and the extinction.

- 4) Figures 4 (schematic of sample flows) and 11 (SMPS plots) are not necessary and could be moved to supplemental information.

We accept the reviewer's suggestion. Figures 4 and 11 were moved to the supplementary material and are now referred to as figures S2 and S3.

- 5) Figure 8 shows good agreement between calculated measured SSA at 404 nm, but both values use the measured extinction. A more direct and convincing demonstration of the agreement would be to compare calculated ($\alpha_{\text{ext}}(404) - \alpha_{\text{sca}}(404)$) and measured ($\alpha_{\text{abs}}(404)$) absorption at 404 nm.

We believe that the reviewer confused our results. We do not calculate both SSA values using the same extinction data points. The **measured** SSA is calculated from direct extinction and absorption measurements measured by the PA-CRD-S. The **retrieved** SSA is calculated from the extrapolated extinction curve that best fits the BBCES extinction measurement (315 to 345 nm and 390 to 420 nm) and the best fitted scattering curve. This is now explicitly explained in the caption of figure 7 that now reads: "Comparison between the retrieved and measured single scattering albedo (SSA) values at 404 nm. The retrieved SSA is calculated from the retrieved extinction and scattering coefficients ($\alpha_{\text{ext}}(t)$ and $\alpha_{\text{sca}}(t)$, respectively), while the measured SSA is calculated from the values of $\alpha_{\text{ext}}(t)$ and $\alpha_{\text{abs}}(t)$ obtained through direct measurement by the single wavelength photo acoustic spectrometer coupled to a cavity ring down aerosol spectrometer (PA-CRD-S)."

- 6) The manuscript would be strengthened significantly by including plots of the extinction/ absorption Angstrom exponents, which could be calculated from the derived spectral values of extinction and absorption.

A plot of extinction and absorption Angstrom coefficient was added to the main text as figure 9.

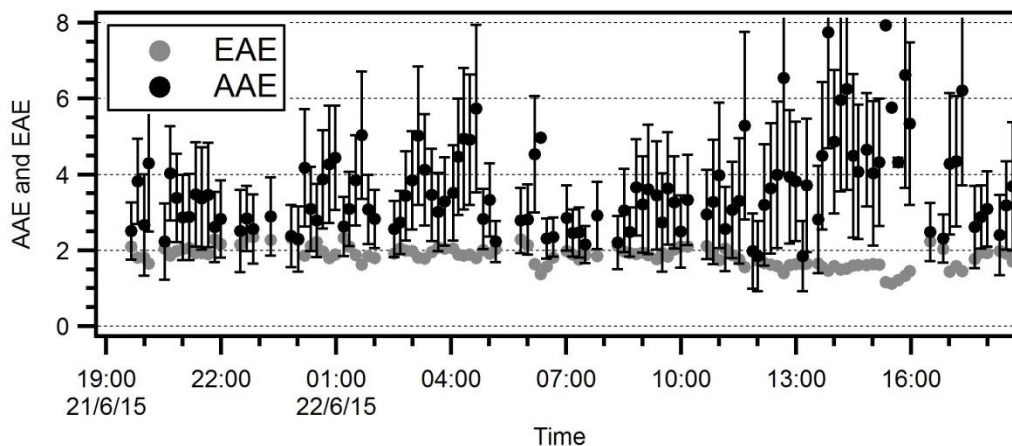


Figure 9. Time series of the retrieved absorption and extinction Angstrom exponents (AAE and EAE) for the 300 to 650 nm wavelength range.

- 7) In the captions for Figures 9 and 10 it would be helpful to remind the reader that these ambient measurements are for the dry aerosol (i.e. do not include the water component which would alter substantially the optical properties). Also, the sampling location should be included in the captions.

We thank the Reviewer for these suggestions. Figure 9 (now, figure 8) caption now reads: “**Figure 9. Time series of the retrieved coefficients for extinction (A), scattering (B), absorption (C), and of the single scattering albedo (SSA) (D) for the 300 to 650 nm wavelength range of dried ambient aerosols.**”

Figure 10 caption now reads “**Figure 10. Time series (night-time hours) of the real and imaginary components of the retrieved effective complex refractive index for the 300 to 650 nm wavelength range of dried ambient aerosols.**”

- 8) To highlight the good agreement between the measured and calculated values, it would be interesting and illustrative to plot curves of the measured extinction (maybe one wavelength for each CES cell), scattering (457 nm, 525 nm, and 637 nm), absorption (404 nm), and SSA (404 nm) in Figures 9a, 9b, 9c, and 9d, respectively.

We refrain from adding additional data to the retrieved optical coefficients figures because it would make these figures too loaded. We would also refrain from adding additional figures to the main text of this manuscript. We believe that figure 8 (retrieved Vs measured SSA at 404 nm, currently figure 7) is sufficient to show the good agreement between measured and calculated values. We did, however, add two additional figures to the supplementary material. One showing the good agreement of the retrieved and measured scattering coefficients at the nephelometer wavelengths (figure S4) and the other showing the agreement of the extinction coefficients at the center wavelengths of the two BBCES cavities (figure S5).

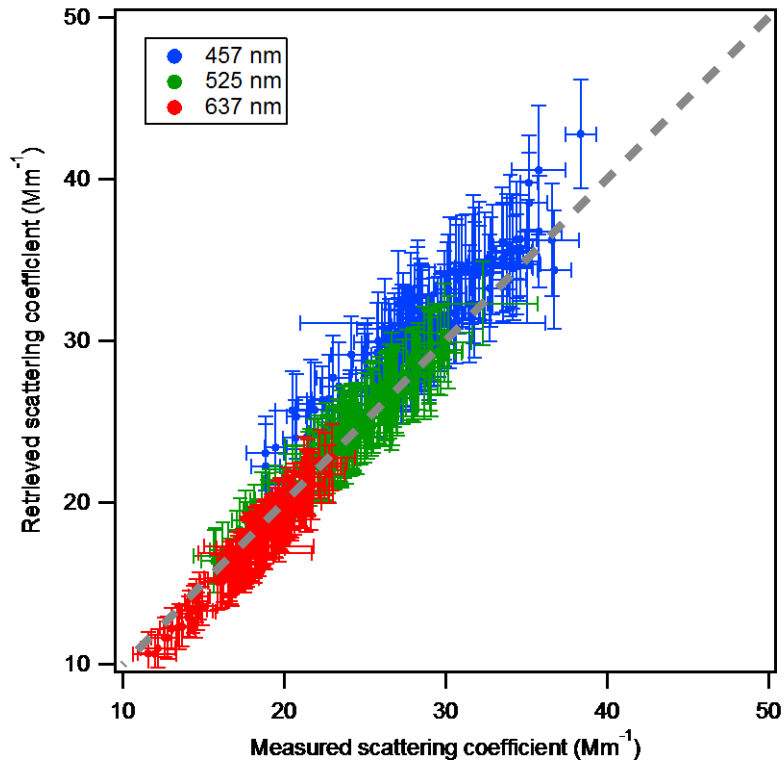


Figure S3. Retrieved and measured scattering coefficients at the nephelometer wavelengths (457, 525 and 637 nm).

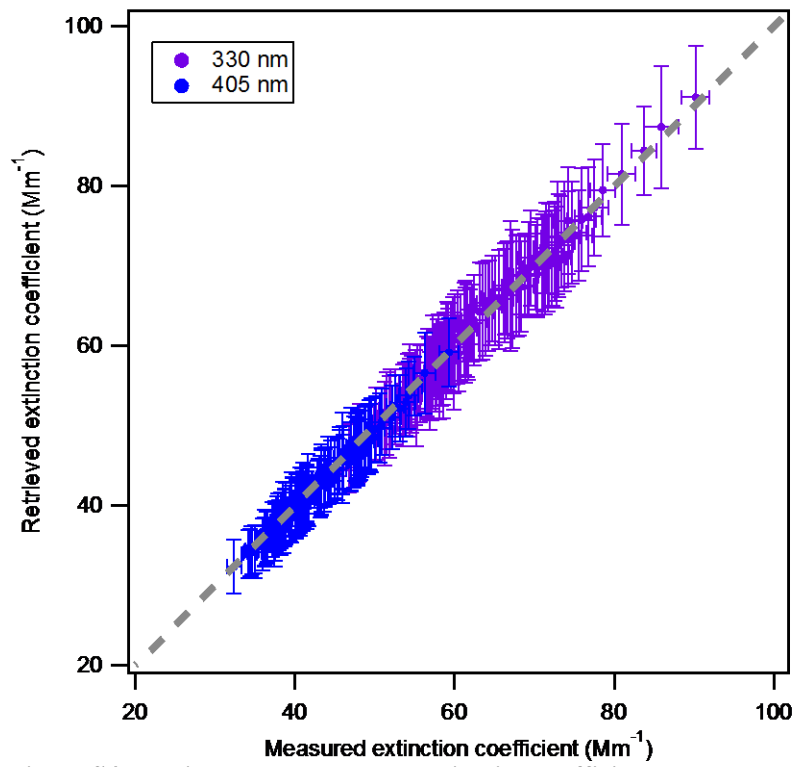


Figure S4. Retrieved and measured extinction coefficients at the center wavelengths of the two BBCES cavities (330 and 405 nm).

- 9) In the Conclusions it is confusing that the authors claim that such a large error on α_{abs} (404 nm) of 60% leads to only negligible errors in total column radiative transfer calculations. And, as the authors point out, the error is expected to be even larger at longer wavelengths where absorption is smaller. It seems like such large errors have to be important given that the absorption extrapolated from 404 nm is used to determine optimum agreement between the calculated and measured scattering values.

In section 3.1. and in the conclusions the value of 60% error at 400 nm (+-60% error is the median error for 400 to 500 nm wavelength range) relates to error of the retrieved absorption coefficient relative to the synthetic data used for computer simulation. It is not an error or an uncertainty on measured absorption coefficients at 404 nm done with the PAS and used to extrapolate absorption data. For clarification, the relevant paragraph in section 3.1. now reads: “Under the conditions of this simulation at 400 nm wavelength (namely; the complex RI, particle size distribution, and number concentration), a relative error of 60% in retrieved values translates into absolute errors of 1 to 3 Mm^{-1} on α_{abs} and of 0.01 to 0.015 on k , respectively.” The relevant conclusion paragraph now reads: “For example, under the conditions of this simulation, at 400 nm, the absolute errors on retrieved α_{abs} and k , are in the range of 1 to 3 Mm^{-1} and 0.01 to 0.015, respectively.”

Technical Corrections

We thank the Reviewer or the careful reading of the manuscript

- 10) Page 1, line 1: “VU-Vis” should read “UV-Vis” **Corrected**
- 11) Page 1, lines 23 and 27: “EFR” should read “ERF” for consistency with other abbreviation (“ERFari”) **Corrected**
- 12) Page 2, line 13: “white-type” should read “White-type” **Corrected**
- 13) Page 2, line 15: the detection limit for the White-type cells should be larger than that of CES, not lower, since they have shorter effective path lengths **Corrected**
- 14) Page 3, line 30: it would seem that the colored glass filters **this comment is not clear, possibly part of it is missing.**
- 15) Page 4, line 28: “flown” should read “flowed” **I think you meant page 5. Corrected**
- 16) Page 7, line 3: “Fig. 2b” should read “Fig. 2a” **Corrected**