

Reply to comments of Reviewer #4:

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) I think this proposed procedure to extrapolate the α_{abs} at 404 nm and α_{ext} at 315-345 nm and 390-450 nm to other wavelength is reasonable only when resonant wavelength of light absorption of particles exist at shorter wavelength compared to these measurement wavelengths. However, some types of SOAs are reported to have maximum light absorption at longer wavelengths. I recommend to adding some discussion on this issue.

We acknowledge the importance of this distinction. The following paragraph was added in the introduction section: “The interaction of atmospheric fine particulate matter with sun light was shown to resemble a power law dependence on wavelength decades ago (Ångström, 1929, 1930, 1961, 1964). The Ångström exponent was since widely used to describe this wavelength dependency and to characterize aerosols size (Valenzuela et al., 2015), composition (Russell et al., 2010) and source (Garg et al., 2016). This monotonic increase in extinction (scattering and absorption) with increasing particle size or with decreasing wavelength is observed for particles with radii smaller than the incident wavelength (or size parameter $< 2\pi$). For larger particles (or shorter wavelengths) the ripple and interference structures of the Mie curves significantly reduce the monotonic increase pattern (Bohren and Huffman, 1983). This also depends on the complex refractive index. Increasing real part limits the power law behavior to smaller particles while increasing imaginary part, dampening the ripple and interference structures, allowing power law behavior for larger particles. Particulate matter with molecular absorption bands in the actinic flux regain would also deviate from the power law spectral dependency. In an ambient dust-free atmosphere as well as in many laboratory and chamber experiments, particles are rarely larger than several hundreds of nanometers making the power law wavelength dependency assumption a reasonable one (Kirchstetter et al., 2004; Hoffer et al., 2006; Sun et al., 2007; Chen and Bond, 2010; Washenfelder et al., 2015).”

- 2) Section 2.2.3: How did you calibrate the IN? How did you estimate the truncation error of the IN? The truncation error should depend on complex refractive index values.

The IN was calibrated by flowing CO₂ and N₂ and relating the measured signal to literature values of the Rayleigh scattering cross sections (Thalman et al., 2014). The truncation angle of the instrument is reported by the manufacturer to be 7-170 degrees (Snider et al., 2015). The truncation error is mostly dependent on the scatterer size and on the scattered wavelength or the particles size parameter. Mie

theory calculation showed that for relatively small particles (up to about 300 nm diameter) the ratio of light intensity scattered at 7-170 degrees to the total scattered light is similar (or not significantly different) to that of Rayleigh scattering regardless of refractive index. In this case the calibration procedure using gasses with known Rayleigh scattering coefficients corrects for the truncation error. When sampling ambient aerosols, mostly composed of primary emissions and SOA with negligible dust component, the concentration of particles larger than 300 nm is negligible. For more details on integrating nephelometer truncation error please refer to Anderson et al., (1996).

- 3) Page 7, line 29: “Both $n(\lambda)$ and $k(\lambda)$ were scaled by two incoherent sine waves to simulate temporal variability.” Could you add some more explanation? How is the amplitude of sine waves? Is it common method to simulate temporal variability? If so, please add a reference.

The reviewer’s question regarding the sine wave amplitude is not clear. The sentence was revised to include the sine waves amplitudes: “Both $n(\lambda)$ and $k(\lambda)$ were scaled by two incoherent sine waves to simulate temporal variability. The sine wave amplitude ranged from 1 to 1.05 for $n(\lambda)$ and from 1 to 1.1 for $k(\lambda)$.”

- 4) Page 8, line 3: The errors in α_{abs} for the calibration of PAS and the truncation correction for IN seem to be larger than 2%.

The uncertainty in absorption coefficient measured with this PAS instrument as a result of the calibration procedure was estimated by Lack et al., (2012) to be less than $\pm 5\%$. The uncertainty in extinction cross section for the BBCES cavities was reported by Washenfelder et al., (2013) to be 3.6-4.1%. With the two largest contributors being the CPC particle counting and the cavity length to filled length ratio (R_L). Both of which are not included in our simulation because we are producing extinction coefficients and not cross sections, and our cavities are built with a central inlet and two sides outlets which reduces the uncertainty in R_L to the uncertainty in length measurement which is about 0.5 mm out of the full cavity length i.e. less than 0.1% . The uncertainty associated with the calibration procedure of the IN is based on uncertainty in temperature and pressure measurements and uncertainty in literature values of Rayleigh scattering cross sections of N_2 and CO_2 . All of which are less than 2% (Thalman et al., 2014). The uncertainty associated to truncation errors of particles that is significantly different from that of the calibration gasses is considered negligible for small particles (Anderson et al., 1996) and is not likely to be larger than 2%. The error we assigned in the simulation is normally distributed with standard deviation of 2%. This means that 68% of the optical coefficients are assigned with errors of up to $\pm 2\%$, 27% of the optical coefficients are assigned with errors between $\pm 2\%$ and $\pm 4\%$ and 4% of the optical coefficients are assigned with errors between $\pm 4\%$ and

±6%. Eventually, based on literature and personal experience we believe that our error assignment is not unreasonable.

- 5) Page 8, line 17: “A complementary N₂ flow of 1.3 SLM was added and mixed with the sample flow, which was then introduced to the IN and subsequently split equally to the SMPS, CPC, PAS, and a three-cavity optical cage that contained the CRD-S and BBCES (see Fig. 4).” Why did you use the N₂ instead of air? I think that the difference may influence to the measurements of PAS, IN, CRD-S, and BBCES.

We respectfully disagree with the reviewer. In the laboratory experiments particles are generated using a particle free dry nitrogen gas (described in the text), so additional nitrogen dilution would not change carrier gas properties.

- 6) Section 3.3 (Ambient aerosol measurement). Many particles (e.g. black carbon (BC) and coated BC, dust) is not homogeneous sphere in the real atmosphere. Some description on this point may be needed.

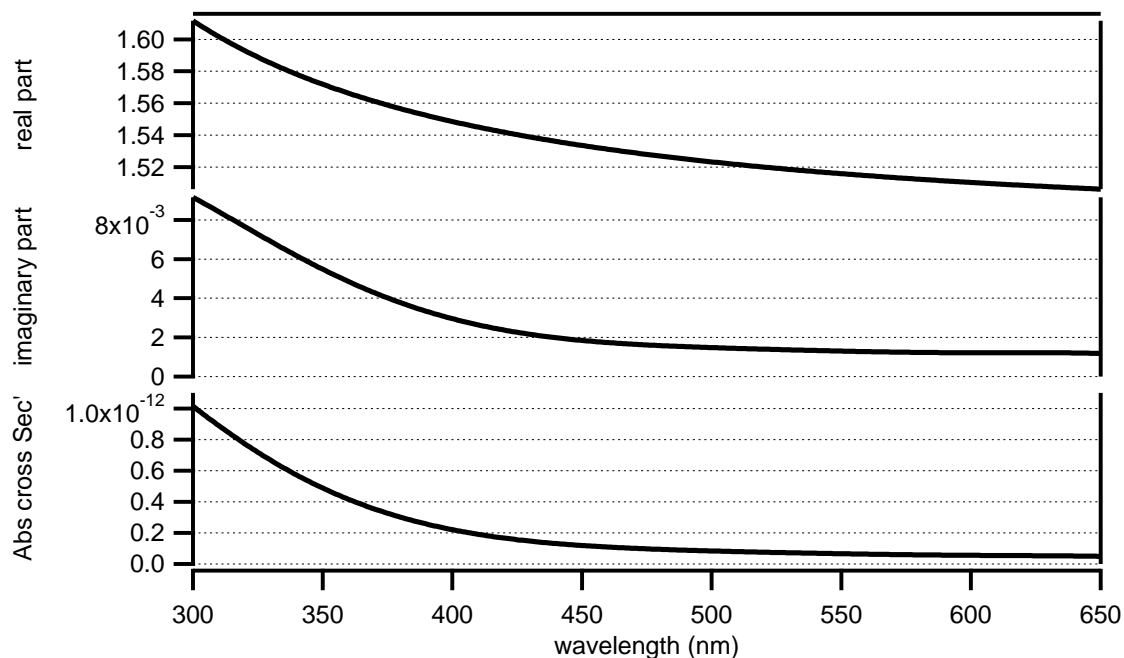
Correct, this is why we defined in the text: “The term effective complex RI is used to represent the whole particle size distribution. It is the complex RI from which, for the corresponding size distribution, we derive the optical coefficients that agree most closely with the measured or input values.” This does not mean that we assume anything regarding the shape, composition and mixing state of the ambient particles.

- 7) Fig. 9(c) The measured and retrieved α_{abs} at 300 nm are more than 10 times larger than α_{abs} at 600 nm throughout the day including the periods when traffic emission seems to have large contributions. Do you think the observed large wavelength dependence (or AAE) is due to the large contributions of brown carbon or inaccuracy of extrapolation of α_{abs} at 404 nm to 600 nm?

A factor of 10 increment in the absorption from 650 nm to 300 nm wavelength would suggest absorption angstrom exponent of about 2.9. Although this is a high value for atmospheric aerosols, values such as these and larger have been reported previously (Hoffer et al., 2004; Kirchstetter et al., 2004; Kaskaoutis et al., 2007; Sun et al., 2007; Chen and Bond, 2010; Russell et al., 2010; Lack et al., 2012; Backman et al., 2014) for BrC aerosols and for desert dust containing aerosol population (which is resendable for east Mediterranean basin during spring). The AAE or the ratio between absorption coefficients at 300 and 650 nm are mostly dependent on the wavelength dependency of the imaginary part of the complex refractive index (RI). Consider the following theoretical example for a single spherical BrC particle with diameter of 80 nm and the effective RI described in the figure. In this example $k_{300}/k_{650}=7.75$ and $\text{Abs}_{300}/\text{Abs}_{650}=20.5$, notice that this theoretical particle is hardly absorbing. We acknowledge the fact that black carbon contribution is expected to reduce the wavelength dependency in an urban

environment. The reviewer's comment led us to find and correct a minor bug in the nephelometer data correction algorithm.

Re-analysis of the measurements showed a stronger decrease in the absorption wavelength dependency (AAE) during the morning hours when higher BC contribution is expected. As can be seen in figures 9c and 9d (now 8c and 8d) there is a general increase in absorption at the longer wavelengths between 07:00 and 10:00 am but it is possible that the nature and distance of the sub-urban to urban sampling site from the main road reduced the black carbon contribution. With the lack of direct ambient aerosols absorption measurements at such wavelengths this question remains an open one.



- 8) Supplemental material. The authors assumed negligible light absorption at 637 nm for ambient particles. I think it not common in urban atmosphere because of the existence of BC. I recommend to adding the validation of the assumption. Unfortunately we cannot show validation of this because we cannot directly measure absorption or extinction minus scattering at 637 nm wavelength. Although this is a very important issue for ambient measurement and for the nephelometer hygroscopic growth correction procedure, it is beside the point for the applicability and robustness of the broadband extrapolation and retrieval procedure because ideally all instrumentation measure the same aerosol population dried or not, as was shown in the laboratory application. Additionally, the fitting procedure of the absorption curve, as described in section 2.3.1 allows for non-zero absorption even at 650 nm. This is also clear from figure 9, showing non-zero retrieved absorption at 650 nm.

Minor comments:

- 9) Page 4, line 1 The value (0.99960) at 330 nm is not consistent with that in Fig.1.
[Thank you. Corrected.](#)
- 10) Page 5, lines 18-19 “The laser power is continuously monitored and used to cancel variations in acoustic signal related to laser power fluctuations.” Where the laser power for the PAS is monitored in Fig. 1?
[This was changed to “The laser power is continuously monitored using a photodiode at the back side mirror, and used to cancel variations in acoustic signal related to laser power fluctuations.”](#)
- 11) Page 7, line 28 I think the “1.856” should be “1.586”. [Revised](#)
- 12) Fig.2(a) I think that one of α_{ext} in the bottom should be α_{abs} . [Revised](#)

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