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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Remote sensing of soot carbon – Part 2: Understanding the absorption Angstrom exponent" *by* G. L. Schuster et al.

G. L. Schuster et al.

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Reviewer 1 comments in bold font, responses in regular font.

The manuscript acp-2015-235 by G.L. Schuster et al. is a well written paper on the limits of using absorption angstrom exponent (AAE) for apportioning the absorption aerosol optical depth (AAOD) to aerosol components (mainly soot carbon-sC, brown carbon-BrC, and mineral) starting from AERONET data. In the reviewer's opinion, the manuscript is very well written, clear, and the dissertation is detailed and coherent. The merit of the paper is to highlight clearly the range of application of the modelling and data analysis performed: They are based on the same hypotheses of the AERONET retrieval algorithm (internally mixed aerosol

with the same complex refractive index for all particles, bimodal size distribution) and cannot be extended e.g. to data directly measured in situ. In the AERONET frame, however, the paper rises important issues commonly neglected in the literature concerning the applicability of the AAE use for AAOD apportionment to aerosol components. The paper shows that under the AERONET retrieval algorithm hypotheses, AAE cannot be used for apportioning the absorption aerosol optical depth (AAOD) to sC assuming that sC $<=>$ AAE=1 as commonly performed in the literature. Modelling was carried out for invariant imaginary k(λ) and the modelled results are supported by real data, which were filtered for invariant imaginary k(λ) and other parameters were used to ensure the presence of different types of aerosols. The results of the paper both show that different AAE for sC can be obtained depending on particle size and sC relative content. Opposite, there exist combination of other components k(λ) (e.g. mineral)/particle size that lead to AAE=1. The reviewer's opinion is that only technical corrections are needed before the paper is accepted for publication.

Thank-you for the thorough read and the constructive comments.

1) Conclusion. It should be clearly stated (as previously extensively done in the text) that the conclusions refer only to the use of AAE calculated from AERONET dataset, as the modelling in the paper is referred to approximations used in the AERONET retrieval process;

Good point. We've changed line 9 of page 20934 to:

"We also noted that particles are necessarily assumed to be internally mixed in the AERONET retrieval algorithm, and that this requires one to interpret the AERONET AAE product differently than one would interpret in situ AAE measurements."

We've also added the following paragraph to the conclusions:

"The analysis presented here mainly applies to the AERONET retrievals, but the scientific community should exercise caution when analyzing in situ data as well. Soot carbon particles that are internally mixed with other aerosol species do not necessarily maintain $AAE = 1$. Additionally, measurement techniques that require dry aerosols may alter the AAE during the drying process (i.e., hygroscopic coatings may evaporate off of sC particles, thereby changing the sC mixing state from internal to external). Finally, although $AAE = 1$ may be suitable for open soot clusters with small primary spherules (radii $\lesssim 0.025\,\mu$m), compact clusters with median effective radii greater than about 0.1 μm may have AAEs close to 0.6; thus, variable sC morphology must also be taken into account when analyzing in situ AAE."

2) equations 4 and 5 illustrate refer to literature approach. Whereas eq. 4 is right for externally mixed aerosol, it is noteworthy that - whereas the reviewer is aware that the approach is commonly used in the literature - eq. 5 is mathematically wrong, as Axy+Bxz cannot be represented as Cxw (i.e. the sum of exponential is not an exponential).

This is an interesting point... Bohren and Clothiaux (2006) devote a section of their book to this topic (Sec 2.1.3), albeit for a different application. They provide a proof with their Eq 2.12, but it is a "proof by contradiction." Thus, they prove that the sum of exponentials is not always an exponential, but they did not prove that the sum of exponentials is never an exponential. They go on to mention that the sum of exponentials is indeed an exponential when their κ is independent of frequency, thereby demonstrating that their section title (i.e., The Sum of Exponentials is not an Exponential) is not universally applicable.

For our own application, we can get an idea of the appropriateness of Equation 5 by putting some numbers into a plotting program. Given the two exponents a and b, we seek to find power law components A and c that satisfy:

$$A\lambda^{-c} = \lambda^{-a} + \lambda^{-b}. \quad (1)$$

If we choose λ =0.3:0.1:1, $a = 2$, $b = 1$, and plot $y = (\lambda^{-2} + \lambda^{-1})$ vs. λ, we get

a linear function in log-log space with a correlation coefficient of 0.9997, a slope of -1.64, and an intercept of 0.292. Thus, $c = 1.64$, $A = 10^{0.292} = 1.957$, and $y = 1.957\lambda^{-1.64}$ is an accurate representation of $\lambda^{-2} + \lambda^{-1}$ (see figure, where the symbols represent $\lambda^{-2} + \lambda^{-1}$ and the line represents $1.957\lambda^{-1.64}$). We repeated this exercise with values of a and b spanning the range of AAE found in the literature, and we always obtain high correlations (see table below). Of course, one could argue that this is not a mathematically rigorous expression (since generally $|R| \neq$ 1.0000), and that would be correct. However, the Angstrom relation itself is not mathematically rigorous either, since it is an empirical expression.

Parameters corresponding to Equation 1.

a	b	c	A	R
1	0.5	0.786	1.988	–0.9999
2	0.5	1.548	1.912	–0.9987
3	0.5	2.470	1.808	–0.9977
1	1.0	1.000	2.000	–1.0000
2	1.0	1.640	1.957	–0.9997
3	1.0	2.496	1.860	–0.9988

3) page 20918 vs. Fig.2 caption: please, be coherent with Rp: 0.026 um is used in the text, 0.025 in the figure caption.
Thank-you. The caption was correct.

4) page 20923: please, define AOD at line 3
Thank-you.

5) page 20928: please check "k=0.36+" at line 23
The value of 0.36 is indeed correct. We changed the text to $k \gtrsim 0.36$ for a more clear presentation.

6) page 20933: please, change "has" into "have" at line 6

Thank-you

page 20934, line 3: double “which" is present
Thank-you

Figure 5: yellow X for coarse median is hardly visible
Thank-you. We have changed the blue diamonds behind the yellow X so that they no longer have white fill. This improves the contrast with the yellow X on our computers; hopefully it is satisfactory on other computers as well.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 20911, 2015.

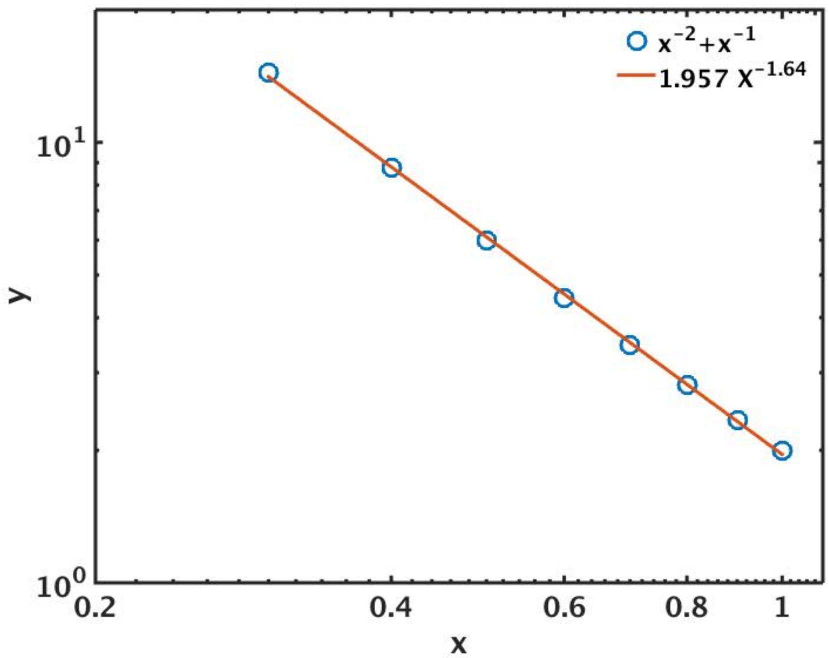

Fig. 1. Plot of Eq 1, with a = 2, b = 1, c = 1.64, and A = 1.957.

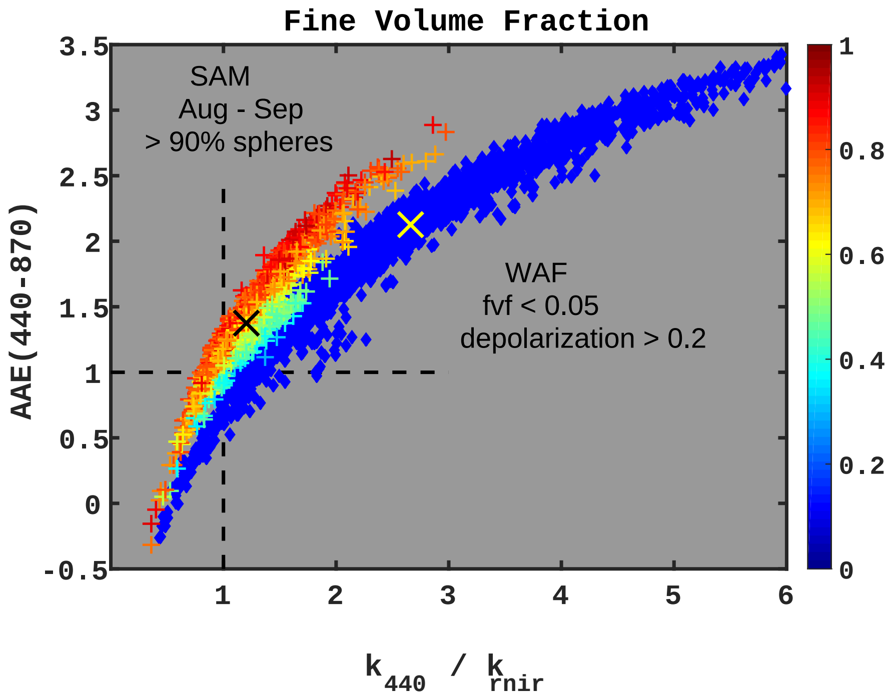

Fig. 2. New Figure 5