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Atmospheric Measurement Techniques Discussions

Interactive comment on "Constrained two-stream algorithm for calculating aerosol light absorption coefficient from the Particle Soot Absorption Photometer" *by* T. Müller et al.

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General comments The paper deals with an important subject. Long-term datasets on aerosol light absorption have been obtained with filter-based methods that suffer from measurements artefacts. The most notable artefacts such as 1) enhanced absorption by multiple scattering and b) the reduction of this enhancement with increasing filter loading and 3) light extinction cause by scattering particles that abusively is interpreted as absorption (apparent absorption) are handled in the manuscript. The authors present a correction method that is new and has a better performance when compared to secondary reference absorption. The correction method is applicable to a

larger range of different aerosol types that are present in the atmosphere. How well the method performs for small particles, e.g. traffic related combustion aerosol, that may penetrate the collecting filter substrate deeper than assumed in the model is yet to be determined. A drawback of the correction method may be that it seems not so straight forward for other PSAP owners to apply the suggested correction method. Making the model/method more readily available would be of great benefit. The essential difference (less loss of information) between the constrained radiation transfer model developed here and a (complex) single formula to obtain the absorption coefficient from filter-based light transmission change measurements remains hidden until page 11112 top-para. I would like to see a paragraph at the end of section 2 that describes why correction formulas can never compete with the model developed in this manuscript. specific comments Abstract. "For high ssa the CTS correction significantly reduces error"...followed by "reduced to 30%." At first reading there may seem to be a contra diction. I suggest to consider reformulation. Section 2 page 11096, line 4. Replace "calculated from the Beer-Lambert law by" by "defined". Beer-Lambert refers to single scatter. Section 2 page 11097, second para on MAAP. Although MAAP is designed so that "No simultaneous measurements of aerosol scattering is needed" please add "additional" before "measurements" and make the remark that remaining cross sensitivity to scattering (apparent absorption) is observed to be within the range 0-3% (Petzold et al., 2005, Mueller et al. 2011). To make the last para of this page in line with this you may consider to higher the remaining cross sensitivity to scattering for PSAP to 2% (650nm) instead of 1%, which is more valid for the shortest PSAP wavelength. Section 2, Last para It is described that solemnly scattering particles lead to apparent absorption and that the apparent absorption becomes a smaller fraction of aerosol light scattering when relative transmittance goes down. Moreover apparent absorption was higher at larger wavelengths. This finding remains unexplained, whereas this may not be so obvious for the reader. Please consider to include a short explanation with moral of story:. "Scattering aerosol, cloud droplets or filter fibers scatter light. Even when the preferred scattering direction is forward, the transmission of light through a

volume with increasing amount of them goes down. In the multiple scattering regime a cloud looks very white from the top a greyish from below (even though every single droplet scatters forward). The higher the optical depth the lower the transmission and for a hypothetical not absorbing cloud the reflection goes up. For the particle collecting filter the same is true. Here I present some calculations to give an impression on how scattering particles are misinterpreted as absorption and how this leads to wavelength dependent apparent absorption. In Arnott et al., 2005 the filter transmission of a PSAP filter is about 22% at 550 nm. The aethalometer filter is physically and optically thicker with pristine filter transmission about 10%. A simple formula is given by equation 14 in the Arnott paper. The equation is valid for hypothetical non absorbing material. From that formula the filter optical depth for PSAP filters is calculated to be about 25, for asymmetry parameter of 0.72. (25 is much higher than the values in this manuscript under review) If now scattering fibers are added on top of the PSAP filter the transmission goes down, the same would be true for aerosol particles. If I use the scattering coefficient shown in Arnott Fig2. (525 Mmˆ-1), I find for the PSAP 0.525 aerosol optical depth (scatter), this leads to an apparent absorption. The same aerosol on an aethalometer filter has obviously less effect as the fractional increase in total optical depth is smaller. Looking at the wavelength dependence of the PSAP filter transmission (pristine), the wavelength dependence is also explained in similar manner. Here, I included a simple figure (fig.1.). Relative transmission is 100% for pristine and 70% for filter change. Filter spotsize and lpm flow taken from Mueller et al, 2011. Asymmetry parameter aerosol and filter same: 0.72. Sensitivity to asymmetry parameter is small if calculated according to equation 4 in this manuscript. For this calculation the apparent absorption is about 3% of scattering coefficient at pristine filter.

The 3% of scattering coefficient is larger than 1-2% and the finding that apparent absorption for aethalometers is smaller than for PSAP (because the change in transmission is relatively smaller as the filter is already optically thick), seems not to be in agreement with the findings of Mueller et al. (2011) but there apparent absorption was already partly corrected for. Section 3 page 11098 line 1-17 Here it becomes some-

what confusing for me. I try to explain: Line 5 “a model (1) is developed”. Line 6 this model includes another model (2) (two-stream RTM) and parametrisations for apparent absorption (this work) and absorption enhancement (Bond and Virkkula). Line 10 “A new correction method was derived from the model for particle-laden filters” I assume derived from model (1)? But the next line the method is given the name CTS thus it is model (2) that is constrained. So I’m confused. Maybe a drawing or scheme may be helpful? Finally CTS, constrained on one side by Bond and Virkkula, is compared to Bond and Virkkula. Final line “one can adopt the correction” what specifically is to be adopted and is “one” the reader or do the authors have plans themselves?. If it is up to the reader, I suggest to include more guidance about Initialisation, constrainments and calibrations. E.g. page 11102 line 6, “illuminated with diffuse light”, is that important when adopting. Section 3.1. It is mentioned that Arnott et al. (2005) used a two stream model. Page 11099 (line 13-14) it is mentioned that in this manuscript “the simpler two-layer model is used” Thus the same modelling approach is chosen and many of the equations following are identical. However what is the difference only becomes clear if the Arnott paper is studied. I would like to read a few lines where this work continues where the work of Arnott et al. stopped. Page 11100 Reasons for the inequality expressed in (3) are given: 1. No independent light scattering.2. Multiple scattering medium. 3.Particle interference. The authors argue that the missing theoretical solutions are solved by empirical parameterizations of apparent absorption and absorption enhancement. To me it is clear how Multiple scattering (2) and absorption enhancement are linked. But I do not see directly how apparent absorption links to the other two. Please comment or just leave it to the conclusion that filter based measurement techniques have artefacts (explained earlier and elsewhere) and that in this manuscript a correction method is developed. Equation 4 and introducing text. Am I confused? The asymmetry parameter can be used to calculate the fraction of light that is scattered forwardly and backwardly. In the single scattering regime this is very convenient. But how does it work with a cloud. The phasefunction of every droplet can properly be integrated to obtain the asymmetry parameter of that drop. However,

for many droplets thus a cloud, all individual droplets with large forward scatter and g close to 1, the cloud becomes highly reflective and the transmission goes down. A dense PSAP filter resembles a cloud and thus although single fibers scatter very strongly in forward direction, g filter is only 0.72 (this work). I wonder what happens if the top filter pack, the borosilicate/glass, is doubled. Obviously the transmission goes down and the asymmetry parameter is getting smaller? So what happens if the filter is loaded with particles can we simply weigh the g filter and g aerosol with the optical depth. I just don't know if equation 4 is applicable here. I do not say it isn't, I cannot judge. Can I calculate g pristine filter form numbers in Figures 5? Page 11106 line 17. Apparently, the factor "mu" is chosen 1/sqrt3 whereas in Moteki it was 1, it is not clear why this is chosen but effectively this difference implies almost a factor of 2 in the total optical depth in equations 6 and 7. "The transmittance calculated from Moteki is an order of magnitude smaller than observations for this study". Choice of the value of mu is suggested as reason, the impact of this choice is easily verified and should be done. Also the asymmetry parameter is suggested as explanation, but Moteki's asymmetry parameter is larger whereas the calculated transmission is smaller, right? Page 11106 line 27. "With this value the calculated enhancement factors are in good agreement with the enhancement factor for the PSAP given in Bond et al." For what experiment or situation? Bond enhancement parametrisation is used as model constrain. How independent is the observation of "good agreement"? Please elaborate a bit on this.

Section 3.1 line 21 "such a model". Many similarities with CTS but what is new here as compared to Arnott 2005 (except applied aethalometer) Page 11109 Do I understand correctly that the calibration experiment for black aerosol is assumed to be not scattering when it comes to sensitivity calculations? Would that mean that some scattering effects are "calibrated-in"? Equations 33 and 34. I cannot rewrite equation 33 in terms of relative optical depth (34) appendix B. Possibly this is related to "that are not yet understood" line 5 page 11114 but I did not check. Page 11115 on the parameterization given. A five(!) parameter fit is compared with the data the fit is based on. The data in Fig 9b seems not to be spread nicely around zero. More importantly the

cases with small asymmetry parameters, thus higher scattering exponents, thus small particles are not well described by the parameterization, i.e. measured 20% too high. A satisfactory explanation for this behavior was not found. Could deeper penetration partly explain the stronger enhancement for small particles?

Typos Start aethalometer with capital A? Page 11094 line 18 "if" replaced by "whether" Page 11108 line 14 "G" must not be capital. Page 11108 limited by?

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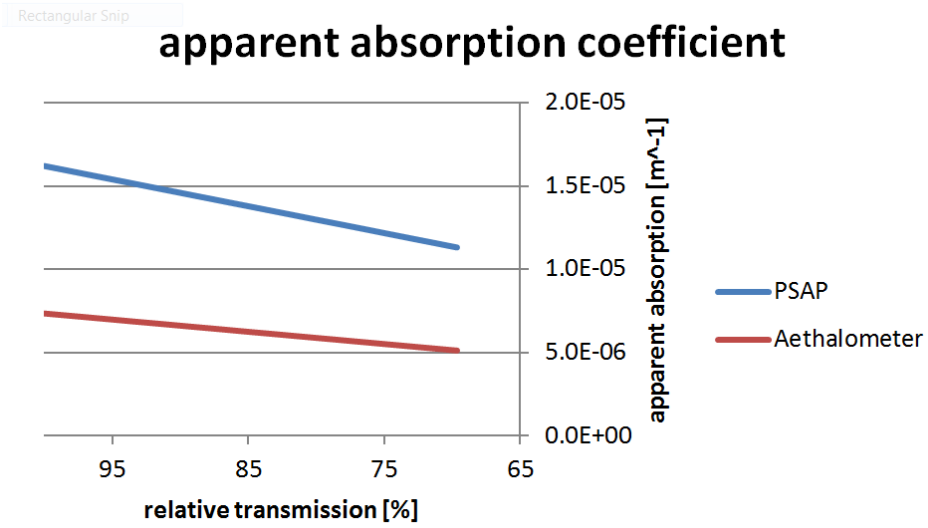
apparent absorption coefficient
2.0E-05
1.5E-05
1.0E-05
5.0E-06
0.0E+00
apparent absorption [m^-1]
PSAP
Aethalometer
95
85
75
65
relative transmission [%]

Fig. 1.