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Atmospheric Measurement Techniques Discussions

Interactive comment on "Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops" *by* T. Müller et al.

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Answer to anonymous Referee #2

The authors want to thank the Referee for the constructive feedback and knowledge to make this article more valuable.

Comment:

The unfortunate conclusion of the paper is that "current correction functions for absorption photometers are not adequate." The authors provide some useful sugges-

tions towards how data quality can be increased (such as regular measuring of spot size and flow). Nonetheless, the results indicate that even when such good practices are put into place (such as at this workshop) significant between instrument variability remains.

Reply:

The authors admit that the message that “current correction functions for absorption photometer are not adequate” is frustrating. There is evidence from other publications (e.g. Nakayama et al. 2010, Lack et al. 2009) that existing correction schemes could be improved or replaced to reduce uncertainties. The Referee claimed in some comments to enhance discussions to get a better insight in possible reasons for uncertainties. The authors will follow the suggestion of the Referee and will revise discussions throughout the manuscript.

Comment:

Additionally, the authors importantly have found that the relative sensitivity of different instruments apparently changed over time when used to sample ambient aerosol. To me, this seems like one of the more important conclusions of the manuscript and I would suggest highlighting it in the abstract, in particular in the context of the robustness of long-term measurements of aerosol light absorption.

Reply:

The authors will mention the changed sensitivity in the abstract of the revised manuscript. A recent publication (Nakayama et al. 2010) showed using monodisperse Nigrosin that the sensitivity of PSAP depends on particle size. The magnitude of this effect with ambient air only can be estimated and is not proven by experiments till now. If Aethalometer or MAAP show a similar size effect is speculation. A discussion of possible changes of the sensitivity due to size effects will be included in the revised manuscript.

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Comment:

Specific questions and comments follow below, but a general comment is that at times I found this paper difficult follow. The paper is comprehensive and very detailed, but I found that the comparison between the measurements from the different instruments was somewhat hard to follow. At times it seemed that unnecessary detail is given while at other times the detail is insufficient to allow the reader to easily follow. Some of this I believe is a result of the PSAP vs. MAAP and aethelometer vs. MAAP discussions becoming too intertwined. My suggestion would be to use further subheadings within sections to highlight the different instrument comparisons. This could be followed with overall summaries which bring the results together (PSAP vs. aethelometer) at the end of each section. Overall, I find this to be an interesting and useful contribution to the field, but I would strongly encourage the authors in their revisions to focus on clarity in their presentation.

Reply:

The authors will consider the suggestions of the referee and remove unnecessary details throughout the manuscript. Additionally the authors will revise some chapters and include subheadings and summaries to improve clarity.

Specific Questions/Comments:

P1521.L17: The likely reason why these workshops did not focus on the effects of organics as they influence filter-based absorption measurements resulted from the timing of the workshops relative to the referenced work. To state that consideration of biases imparted by organics or other “liquid” aerosol was far beyond the scope of the workshops does not seem appropriate. Understanding such biases in detail is critical to quantitatively understanding the performance of filter-based absorption photometers in the real world. Without such knowledge, the quality of the measurements cannot be assured for anything but the test aerosols considered here. For example, a goal of these workshops is to establish methods to ensure high quality data that are compa-

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rable across many different sites. These sites may be impacted by aerosol with very different compositions and therefore with different associated biases.

Reply:

The authors will replace that sentence by: 'Although it is important to understand biases in filter based measurements due to organics and liquids, dedicated experiments concerning the response to organics were not performed during the workshops. To achieve tasks I to IV it was chosen to use well defined aerosols and not to generate aerosols with large amount of organics or liquids."

Comment:

P1525.L1: I recommend breaking this into three paragraphs where each paragraph deals with each instrument type in turn.

Reply:

The authors will break it into three paragraph.

Comment:

P1530.L7: The calculation of "R" appears to require knowledge of the single scatter albedo. Barring the use of other instruments, how is the SSA generally determined for use in this calculation? The SSA cannot be known until the absorption is known, but the absorption measurement depends on the SSA. Thus, the correction appears to be circular. For the purposes of this manuscript, it is stated that "All Aethelometers are corrected by the same experiment/measurement-period average f value." From which data is the average f determined? The MAAP + PSAP?

Reply:

The value f-values were determined using the absorption coefficient measured with MAAP and scattering coefficient measured with nephelometer. To adjust the measured scattering and absorption to other wavelengths, the Ångström exponents for scattering

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and absorption were used. Ångström exponents for absorption were derived from corrected Bond et al. (1999) corrected PSAP values. Thus aethalometer was not used as a stand-alone instrument, but this approach allows to estimate an f-value without cicular reference. The authors will add this information to the revised manuscript.

Comment:

P1531.Footnote 2: It is not abundantly clear what is meant by this footnote.

Reply:

The authors will remove the footnote and change the text to correct for an additional error in the serial numbers: "Several types of PSAPs (Radiance Research) were tested during both workshops. Instruments with serial number 13, 15, 20A, 20B, 28, 48, and 60 had a peak of the light emission at wavelength 565 nm. A tail of the emitted radiation at longer wavelengths causes an effective wavelength of about 585 nm. For a typical wavelength dependence of lambdaˆ-1 the ratio of absorption coefficients at 565 and 585 nm is 1.035. A newer instrument with serial number 71 had a symmetrical intensity distribution with FWHM of 20 nm and a peak wavelength of 522 nm. The three-wavelength PSAPs (serial number equal or larger 80) show peak wavelengths.. . ."

Comment:

P1532.L12: It is not clear whether the Angstrom exponents were determined from wavelength pairs (i.e. following from Equation 1) or from a fit to all three wavelength points. These two methods can give somewhat different results so it should be stated more explicitly how this was done.

Reply:

The Ångstom exponent was the average of two wavelength pairs. The authors will clarify this in the revised manuscript.

Comment:

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P1533.L6: It is stated that the increasing standard deviation in the PSAP spot area measurements can be explained by “bad sealing rings.” It is not abundantly clear why this would have a greater influence on the larger area spots.

Reply:

We agree that the explanation for the correlation between spot area and standard deviation is ill-founded and a speculation. We will delete the respective sentence.

Comment:

Related to this, it is not clear whether the actual spot (i.e. the cut out portion of the filter holder) or the spot produced on the filter from aerosol deposition was actually measured.

Reply:

It is always referred to the actually measured spot produced on the filter. The authors will change the sentence (page 1532, line 20) to: “During both workshops spot areas produced on the filter were measured with optical reticles. . .”

Comment:

P1533.L24: Can the authors comment on why the average spot size would be lower during one workshop than the other? Did something in the design of the aethelometers change?

Reply:

The authors will add following sentence (page 1533, line 26). “There is no explanation for differences among both workshops. Measuring the spot size with a caliper might be subjective, but there is no evidence that this is the only reason for differences among both workshops. Changes of the design of Aethalometer between the workshops are excluded to be responsible for differences.”

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Comment:

P1535.L26: I find much of Section 5.3 difficult to follow, in particular the definition of the "face factor" and the associated discussion. It is somewhat difficult to really understand what is being compared to what. Perhaps a table summarizing the comparisons would help (although there are already a lot of tables)?

Reply:

The authors will revise section 5.3. The description of the procedure will be tightened and the presentation of results will be improved.

Comment:

Figure 3: Given the log scale, it is not clear why the ambient measurements needed to be multiplied by a factor of 10 for presentation. I suggest presenting the unscaled data to avoid confusion and give a more direct representation of the actual conditions.

Reply:

The authors will follow the suggestion of the referee and shown unscaled data.

Comment:

P1537.L21. How was the noise/time relationship actually determined from the experiments? It is not clear.

Reply: There is a mistake that chapter. The relationship $\Delta t\hat{}$-1.3 and was derived not in this manuscript but in Sedlacek et al. (2007). The time/noise relationship was not determined by the authors. The passage will be changed to: "In contrast, experiments done by Sedlac et al. (2007) showed that the noise of the PSAP varies with $\Delta t\hat{}-1.3$ and the noise was determined to be 1.6 Mm$\hat{}-1$ for an averaging time of 2 s. Using this noise time relationship the noise of PSAP should be 0.02 Mm$\hat{}-1$ for an averaging time of 60 s. However, the lowest values for instrumental noise at averaging time of 60 s found during the GAW2005 and EUSAAR2007 workshops are about 0.05 Mm$\hat{}$-1 and

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thus 2.5 times higher than the theoretical value."

Comment:

Section 5.4: It needs to be made clear whether the authors are reporting standard deviations or standard deviations of the mean. How long was data collected for?

Reply:

The authors mean standard deviation. The sampling time of the time series varied between 80 and 150 min. The authors will clarify this issue and change the first paragraph of chapter 5.4: ". . .. Averaging times were set to one minute for PSAP and MAAP, and three minutes for Aethalometer. The instrumental noise is defined as the single standard deviation of absorption coefficients collected during the sampling period, and thus the instrumental noise is the uncertainty of a single readout when sampling filtered air. Typically the detection limit of instruments is defined as three times the instrumental noise. The instrumental noise was determined for each instrument. Averages, maximum and minimum noise of instruments of the same type are given in Table 9."

Comment:

As with Section 5.3, I find the discussion in this section to be somewhat difficult to follow. A better description of the experiment and the procedures used would be helpful.

Reply:

The authors agree to the referees objections. The section will be revised.

Comment:

P1539.L10: The need to convert the standard deviations to face velocity equivalent values is not overly clear. I would think that what is more important is the standard deviation under what could be considered "typical operating conditions," which could be determined from Section 5.3.

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Reply:

The authors think that is interesting to test if the noise and face velocity are inversely proportional as proposed by Springston and Sedlacek (2007). But we agree that it is more important to give the noise at typical operating conditions. The authors will change the text with focus on typical operating conditions.

Comment:

P1541.L8: A value of 0.79 is given here, but 0.81 is given in Table 10.

Reply:

The correct value is 0.79. The authors will correct Table 10.

Comment:

Figure 9: Why is it that all of the high absorption level data are at “high loading” while the low absorption data are at “low loading”? Upon first glance, the figure appears to show a major deviation between the PSAP and MAAP at high absorption levels, but presumably this is just because of the filter transmittance differences. Were there any periods where the absorption was high and filter loading low (and vice versa)? Also, it seems as if the fits have been forced through zero (although this was not stated). If the fits are not forced through zero then the discrepancy at high loadings will appear to be greater in terms of the slope and the intercept will be significant.

Reply:

The ambient air runs typically were started in the evening and last until morning of the next day. Then the transmittance is low in the early morning hours and higher soot concentrations caused by the morning rush hour are correlated with high loadings. We haven’t found data where the absorption is high and the loading is low (and vice versa). The fits are forced through zero. The authors will state it in the text. The authors think that a linear fit with slope and intercept is not meaningful. If a fit is not forced

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through zero a significant intercept can occur due a nonlinear relation between both instruments, although the track of points can hit the origin for low absorption coefficients (within the uncertainties of instruments). Thus the authors think that a fit forced through zero is more meaningful. A fit not forced through zero can be physically meaningful for instruments with drifting background, e.g. photoacoustic photometers, extinction cells or nephelometers. This is not the case for filter based absorption photometers.

Comment:

Figure 10: Again, was the fit forced through zero? It appears to be so, but this is not stated. What is the slope/intercept if the fits are not forced through zero?

Reply:

Again, the authors think that a fit not forced through zero are not meaningful. Additionally, a discussion including more than one fitting method complicates the presentation of results. The authors will mention in the revised manuscript that the fit was forced through zero.

Comment:

P1543.L12: It should be clarified that the relative sensitivities are relative to the MAAP measurements.

Reply:

The authors will state that the relative sensitivity are relative to MAAP.

Comment:

P1543.L10-21: The authors have found that relative sensitivities differ between campaigns. This suggests that the sensitivity may be a function of the ambient aerosol composition. As such, I'll pose the question that if the instruments are really this sensitive to the specific conditions of the ambient aerosol, then is there really any hope of obtaining accurate absorption measurements over long periods of time at different

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sites? How could one be certain that observed trends are actually a result of actual changes in aerosol absorption vs. changes in aerosol composition unrelated to absorption? I would suggest some discussion of this be included.

Reply:

There is evidence that filter based measurements are affected by more parameters than the parameters used in the corrections given in e.g. Bond et al. (1999) and Weingartner et al.(2003). Figures 12 and 13 show that there is the need to improve the scattering corrections. Recently it was shown by Nakayama et al. (2010) that the sensitivity to absorbing particles depends on the particle size. Thus there are more parameter controlling the sensitivity which are not accounted for in Bond et al. (1999) and Weingartner et al.(2003). Further experiments are needed to develop more sophisticated corrections. The authors will include this issues in the revised manuscript.

Comment:

P1545.L13: The upper value given here is only 2.18%, but the largest value in the table is 3.12%. In the table it is indicated that this upper value has been excluded for some reason, but this is not explained.

Reply:

The largest value of 3.12% was measured with PSAP 013. Because of a weak light source the noise of that PSAP was about four this higher compared to the other PSAPs. Data of PSAP 013 were not used for any data evaluation. To avoid confusion the authors will delete PSAP 013 from Table 11.

Comment:

P1546.L15 and P1547.L6 and Figure 12: Can the authors comment on why the scattering effect appears to level off at high filter loadings?

Reply:

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The authors regret that they cannot explain this observation with data presented in this manuscript. An explanation would require radiative transfer calculations to simulate the optical properties of particle laden filter, what is far beyond the scope of this manuscript.

Comment:

P1547.L13: Do the authors have reason to believe that in previous studies investigators would have used “dirty” filters at the start of their experiments? Certainly, this would have been indicated if it were the case. Given that one of the authors on this paper was also an author on the RAOS study paper it would seem that there should be direct knowledge of whether this was the case.

Reply:

It is not the intention of the authors to blame other investigator to have done their experiments without the necessary care. Investigations concerning the sensitivity to purely scattering particles are very sensitive to the transmittance. It is easy to see (c.f. Fig 12) that the sensitivity to purely scattering particles is very sensitive to the transmittance. User of PSAP know that the transmittance can change rapidly after inserting a new filter even if the instrument samples particle free air. It can take few minutes to have stable conditions and the transmittance can be set manually to unity. This is not an issue for monitoring purposes, but experiments similar to that one shown in Figure 12 are very sensitive to transmittance. If high concentrations of scattering particles are sampled while changing the filter, a significant amount of particles can be sample on the filter until the transmittance was set to unity. The authors worked out these findings after the second workshop. Thus data presented in this manuscript also suffer from an undefined “preloading” of filters. The authors will change the text (P1547.L13) to.

“It is hard to compare results from different experiments if there could be an even small preloading before starting the experiments. The authors worked out these findings after the second workshop. Thus also data presented in this manuscript suffer from an

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undefined preloading of filters which can introduce large uncertainties in the results. Thus it would be desirable to perform experiments taking into account the problems with preloaded filters."

Comment:

P1548.L8: I believe that the MAAP applies an internal correction. Additionally, the MAAP absorption values were all increased by 5% following comparison with a photoacoustic instrument. Thus, it appears that an empirical correction for the MAAP was actually applied.

Reply:

The authors will correct a mistake, since MAAP data were increased by 5% to account for the wavelength adjustments as shown in line 14.

Comment:

Table 10 vs. Table 11: It is interesting that the aethelometer/MAAP ratio for the ambient aerosol during GAW2005 is only 1.37 while for carbon black it is 2.11 and 1.53 for the two aethelometers reported. Can the authors offer a reason for the dramatically different behavior of the ambient vs. carbon black results? The findings of Lack et al. (2008) (which suggest that OC may bias filter-based absorption measurements) would suggest that, if anything, the deviation in the ambient observations would be expected to be greater than for the carbon black experiments.

Reply:

As mentioned before, there are more parameter controlling the sensitivity of filter based measurements, e.g. the dependence of particle size (c.f. Nakayama et al. 2010). Smaller particles are deposited deeper in the filter what increases the sensitivity. For PSAP the ratio of sensitivities for particles sizes of 100 nm and 200 nm is about 1.6! That certainly can not explain the differences of about 38% between the two aethalometers during the GAW2005 workshop. The authors will add a section to

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the discussion to address the differences when comparing different workshops and aerosol. A discussion of size effects and a possible bias due to OC will be included.

Comment:

General: the authors spend a lot of time going over the results of the ambient aerosol observations and the standard deviations associated with these measurements. However, when it comes to the more constrained Carbon Black measurements the discussion is significantly less. Wouldn't the laboratory experiments with Carbon Black be better suited for detailed analysis as this takes out some of the uncertainty regarding aerosol composition? The conclusion that "differences between the absorption measured by the MAAP, PSAP and aethalometer cannot be explained" is unsatisfying and I suggest should be discussed further, particularly in light of the ambient measurements. Do the results from the carbon black studies suggest that the level of agreement between the different instrument types is worse than indicated from the ambient measurements? At minimum, I believe that the results from the carbon black studies should be mentioned in the conclusions alongside the ambient aerosol results.

Reply:

The outcome of the workshops is that sensitivities of filter based absorption photometers to different absorbing aerosol are not well understood. Sensitivities may depend on chemical composition and more partly unknown parameters. Thus laboratory experiments with a better characterized aerosol (e.g. carbon black ammonium sulfate) are important for understanding the fundamentals filter based absorption photometers. Finally results of laboratory experiments should lead to better correction schemes, which to improve measurements of ambient air.

The authors agree to the referees objections and will enlarge chapter 6.4 (Instrument intercomparison with black particle) in the revised manuscript. Also the authors will upgrade the conclusion. Differences in the relative sensitivities with ambient air and black carbon will be discussed in the conclusions.

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Technical Corrections:

Table 2: No values are reported for Carbon Black.

Reply:

Table 2 reports values of the EUSAAR2007 workshop. Data for carbon black experiments are only available for the GAW2005 workshop (Table 1).

Comment:

P1529.L15: Berkeley is mis-spelled.

Reply: Will be corrected.

Comment:

Equation 14: This equation has sigma_*_abs, but the definition following uses sigma_*_ap.

Reply:

The authors will correct equation 14.

References not found in the discussion paper:

Lack, D. A., Cappa, C. D., Cross, E. S., Massoli, P., Ahern, A. T., Davidovits, P., and Onasch, T. B.: Absorption Enhancement of Coated Absorbing Aerosols: Validation of the Photo-Acoustic Technique for Measuring the Enhancement Aerosol Science and Technology, 43, 1006-1012, 2009.

Nakayama, T., Kondo, Y., Moteki, N., Sahu, L. K., Kinase, T., Kita, K. and Matsumi, Y. (2010). Size-dependent correction factors for absorption measurements using filter-based photometers: PSAP and COSMOS. Journal of Aerosol Science 41:333-343.

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 1511, 2010.