Response to review – RC2

General informations:

Changes in the manuscript are highlighted with dark red. *Italic* written text is the citation of the reviewers. Blue written text is the response of the authors. Longer text passages additionally included in the manuscript are highlighted with red. The revised version of the manuscript will be uploaded.

Review of anonymous Referee #4

Remark: „ *The paper describes the effects of variable humidity on measurements of light absorption with filter-based absorption photometers. It has been known for a long time that elevated relative humidity distorts these measurements and this is the main reason why it has been recommended to only use data sampled at dry conditions. In certain type of measurements, especially recently popular balloon or drone-borne vertical measurements drying is not possible and rh is rapidly changing. It is therefore important to characterize the responses of the absorption photometers to the changing conditions. This is what this paper presents. It is probably the first one that actually quantifies the effect.*

It is an interesting piece of work. For me the most interesting observation was the completely opposite response of the two absorption photometers and actually of the different filter materials. This very interesting indeed. Actually, it should be emphasized in the conclusions that the responses are filter material dependend and should be quantified if and when new filter materials are used in whatever filter-based absorption photometer, not just the two instruments used in this work. I can recommend the publication of the paper in AMT after some modifications. I did not find any major errors even though I did find some relatively small points to correct and change to the revised version of the ms. I will list them in the detailed comments below:"

Response:

We thank for the review. We will consider the general remark and emphasize, that it is a crucial task to quantify the effect of any absorption photometer, either by following our experimental setup or by other similar approaches. Especially since the STAP can operate with different filter material. Furthermore, more measurements regarding the MA200 have to be conducted to fully understand all the processes. In the conclusion is already stated that the opposing behavior is caused by the different filter material. As recommendation we added: "5. Since the response is different in magnitude and sign for both filter materials, we recommend to examine the effect for other filter materials as well."

The reviewer provided more comments and ongoing we will address them point by point:

Key point 1: *"L106. "Ogren (2010) published the loading correction ..." and then the Eq. (4) is shown. This is not quite correct. Ogren (2010) presented a corrected version of the equation which was originally presented and also corrected by Bond et al. (1999)."*

Response:

Thanks for that comment. We think the reviewer means that the sentence itself is wrong, and we update to: "For instance, Ogren (2010) reported an updated loading correction function for the PSAP introduced and updated by Bond et al. (1999) defined as:". We directly took the Eq. from the manual of the STAP so the Eq. (4) should be mentioned, since it is used internally.

Key point 2: *"L124-126 "... we used the σ_{abs} directly provided by the STAP and derived with the mentioned MAC in the case for the MA200, which already accounts for multiple scattering and filter loading corrections." How does MA200 account for multiple scattering and filter loading? What function is valid for Teflon? For STAP they are probably assumed to be done with the multiplication by Eq. (4), right?"*

Response:

Thanks for that comment. Yes, for the STAP the correction function from Eq. (4) is used internally by the STAP itself to account for filter loading effects. In the manual of the MA200 there is no loading correction function given. Anyhow, Jimenez et al. (2007) empirically determined a loading correction function *K(ATN)* for the Teflon filter-based AE41 and AE42 (Magee Scientific Company). This equation corrects the measured black carbon mass concentration M_{BC} as follows: $M_{BC} = \dfrac{b_{ATN}}{\sigma_{ATN}} \times \dfrac{1}{K(ATN)}$,

in which b_{ATN} is the attenuation coefficient in m^{-1} and σ_{ATN} the attenuation efficiency of BC with a value of 16.6 $m^2\,g^{-1}$ at 880 nm. *K(ATN)* is defined as:

$$K(ATN) = a + b \times \exp(\frac{-ATN}{100}),$$

with *a* and *b* some linear regression coefficients. Jimenez at al. (2007) reported values for *a* and *b* of 0.13 and 0.88. Besides that correction function, which could be applied during post-processing, the MA200 features a DualSpot© loading correction approach. Another Aethalometer (AE33) also uses the dual spot approach. A comprehensive correction approach using light attenuation measurements of two sample spots is provided by Drinovec et al. (2015). But, since the DualSpot© mode was not working during the time of the experiment, this also cannot be applied to the given data set. Hence, we suspect that there was no loading correction applied internally to the measurements of the MA200. Furthermore, since we do not know, if the MA200 needs the same loading correction of the AE41 or AE42, we did not consider any loading correction and decided to use the data directly measured by the instrument. This, if so, should only influence the results by a view per cent. As shown in the given manuscript, the areal loading density as well as the loading material do not change the response of the MA200 to *rh* changes. Therefore, we think a loading correction is negligible.

References: Jorge Jimenez, Candis Claiborn, Timothy Larson, Timothy Gould, ThomasW. Kirchstetter & Lara Gundel (2007) Loading Effect Correction for Real-Time AethalometerMeasurements of Fresh Diesel

Soot, Journal of the Air & Waste Management Association, 57:7,868-873, DOI: 10.3155/1047-3289.57.7.868

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A., and Hansen, A. D. A.: The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8, 1965-1979, https://doi.org/10.5194/amt-8-1965-2015, 2015.

Key point 3: *"2) The manufacturers, their addresses, and filter materials used in the photometers are presented on lines 79-82, lines 129-132, lines 135-137, and 147-150. Maybe once would be enough."*

Response:

Thanks for that comment. Yes, we totally agree, that mentioning once is enough. Therefore, we removed the first, second and fourth mentioning and kept the manufactures addresses in the instrument description part. The used filter materials are important, therefore we think it is worth to repeat the used filter material several times to emphasize that.

Key point 4: *"L140. The reference to Holder et al. (2018) is to a conference abstract. I checked it at the conference book of abstracts. Sure, the abstract is there but it is so short that it does not contain any of the information you write on lines 139-145. If you cannot find anything that can be checked by a reader, you should remove these lines."*

Response:

Thanks for that comment. The given reference refers to a poster presented during the 10th IAC in St. Louis, MO, USA. We updated the reference. We included the url which links to the webpage where the poster can be downloaded. This contains all the given information mentioned in the manuscript.

Holder, A., B. Seay, A. Brashear, T. Yelverton, J. Blair, and S. Blair. Evaluation of a multi-wavelength black carbon sensor, Poster, 10th International Aerosol Conference, St. Louis, MO, September 02 - 07, url: https://cfpub.epa.gov/si/si_public_record_report.cfm?Lab=NRMRL&dirEntryId=342614, 2018.

Key point 5: *"L213-214 "... Filter loading mass is calculated by multiplying the apparent loading mass concentration of the considered material..." What is apparent loading mass? Define it. Where do you get it from?"*

Response:

Thanks for that comment. We think we did not used the correct words to explain what we meant. We therefor reworded the 5th sentence in Section 3.2.: "For both considered loading materials, the mass loaded onto the filters was calculated by multiplying the prevalent loading mass concentration within the mixing chamber with the volume flow rate of the instrument and the loading duration.". The calculation of the prevalent loading mass concentration within the chamber is explained for both loading materials in the beginning of the Section 3.2.1 and 3.2.2 separately. For ammonium sulfate the ammonium sulfate

volume concentration (integral of the volume size distribution) within the chamber was multiplied an assumed ammonium sulfate density of 1.77 g/cm^{-3}. For BC different approaches had to be considered. We added: "During experiment #1 the mean absorption coefficient of the STAP was divided by a *MAC* of 6.6 m^2 g^{-1} since the absorption was stable during the loading period and it's a direct measure from the sampling instrument. For the experiment #2 the loading mass concentration was taken from the average of two consecutive MAAP measurements since the loading period was shorter than 2 minutes which is shorter than the internal averaging period of the STAP so that no stable absorption coefficient readouts could be provided by the STAP. During experiment #3 no MAAP was available and the absorption coefficient measured by the STAP was unstable. We therefore decided to estimate the loaded eBC mass by integrating the absorption coefficient during the loading period and dividing it by the *MAC*.". This should clarify how the loadings on the filters were derived. Furthermore, we think that the word "apparent" is wrong it that context.

Key point 6: "*L221-222 " Four different ρ* were .. for STAP, three for the MA200 ...". Were they not sampling simultaneously?*"

Response:

Thanks for that comment. For both instruments the first three filter areal loading densities were estimated simultaneously. Because of the lower flow rate and the different spot sizes these densities are different. The fourth loading density in the case of the BC was only estimated for the STAP since no MA200 was available during this time. Anyhow, the loading density is in the same range of previous loading periods and therefore at least on case with that areal loading density is covered. The MA200 was also not available during another ammonium sulfate experiment and therefore we have considered 11 areal loading densities for the STAP and 8 for the MA200 in this case. We changed the third last sentence in Section 2.3 to: "The loading aerosol was split into two streams from one of which the absorption photometer were sampling simultaneously." to emphasize that both photometer were sampling the same aerosol.

Key point 7: "*When I look at fig 5 I see that the time when ammonium sulfate was sampled was hours. How stable could you keep the AS production? How would possible instabilities affect the result?*"

Response:

Thanks for that comment. We consider, that we missed to explicitly point out that in the experiments considering different areal loading densities, the filter was loaded before the *rh* was changed. We therefore added the sentence:" The filter were loaded to a certain extent with different materials and afterwards the absorption photometer were sampling particle free air with adjustable humidity." In Section 3.2. Also we added:" Two main setups were used to investigate the effect of changes in real humidity. In the first the filters of the devices were unloaded and the instruments collected a particle free airflow with adjustable relative humidity. In the second, the filters of the devices are loaded to a certain degree and afterwards they sample particle-free humidified air." as the second last paragraph in Section 2.3.

The loading periods lasted 20 minutes at most. During the loading periods we generated ammonium sulfate with an atomizer, which produces very stable loading mass concentrations (narrow standard

deviation around the mean particle number and volume size distribution in Figure 2) when the aerosol chamber is well-mixed. Only during the well-mixed states of the mixing-chamber the filters were loaded. We added: "The very narrow standard deviation around the mean particle number and volume size distribution in **Fehler! Verweisquelle konnte nicht gefunden werden.** indicate clearly that the loading mass concentrations were very stable during the loading periods." at the end of the first paragraph in Section 3.2.2.

Key point 8: *"L257-261. There is speculation about possible effects of the negligibly small imaginary index of AS. There is a more plausible explanation. Why wouldn't the explanation be the apparent absorption or cross sensitivity of any filter-based absorption photometer to purely scattering aerosol that the authors are well aware of? The apparent absorption should be mentioned and discussed at some point of the paper already earlier."*

Response:

Thanks for that comment. This part is speculative and we removed the very negligibly small imaginary part of ammonium sulfate as a possible explanation. We agree that the cross-sensitivity of the absorption photometer to purely scattering aerosol is more important. The problem is, that the sensitivity to ammonium sulfate (Fig 6., measuring absorption during the loading period around 18:30 and 21:00 UTC) is also visible for the MA200 and it is not showing any variation in the response to relative humidity changes across different loading materials which could mean that PTFE membrane filter is unaffected by filter loading in terms of the *rh* effect or the loading was too low. This has to be tested in further studys. Anyhow, we updated the 5th paragraph in Section 3.2.2 to: "As shown in Fig. 6, both absorption photometers measure an "apparent" absorption coefficient of approximately 2 Mm^{-1} during loading with ammonium sulfate (18:30 and 21:00 UTC). This shows that absorption photometers react sensitively to scattering aerosols such as ammonium sulfate. The scattering ability of any material can be described with the real part of its refractive index. It seems that for the STAP the slope of the correlation increases with increasing scattering of the loading material (0.15 Mm^{-1} %$^{-1}$ for a clean filter, 0.21 Mm^{-1} %$^{-1}$ for ammonium sulfate, and 0.30 Mm^{-1} %$^{-1}$ for BC). Ammonium sulfate has a real part of 1.521 ± 0.002 (at 532 nm Dinar et al, 2007) and BC from combustion processes has a real part of 1.96 at 530 nm (Kim et al., 2015 following Ackermann and Toon (1981)). Hence, the quartz fiber glass filters loaded with "artificially" absorbing aerosol inside the STAP could lead to a variation in the response to relative humidity changes. But, the MA200 was loaded with ammonium sulfate as well and its response to relative humidity changes is almost constant for all considered loading materials. Therefore, either the observation is caused by the interaction of quartz fiber glass filters with the loading material and the PTFE filter inside the MA200 do not causes this behavior, the filter loading of the MA200 was too low, or there are other mechanisms explaining this. Furthermore, since only three different cases (clean, ammonium sulfate and BC) were observed in this study more materials should be considered to investigate this phenomenon.".

Key point 9: *"Section 3.3. This section contains no other information but that a dryer dries humid air. The points in fig 7 are on the same line with and without drying so it does not tell anything about the responses of the absorption photometers. You would have obtained the same points also by reducing the original humidity. Even hypothetically there should not be a difference in reducing the original humidity or reducing it afterwards with a drier. Just omit the section."*
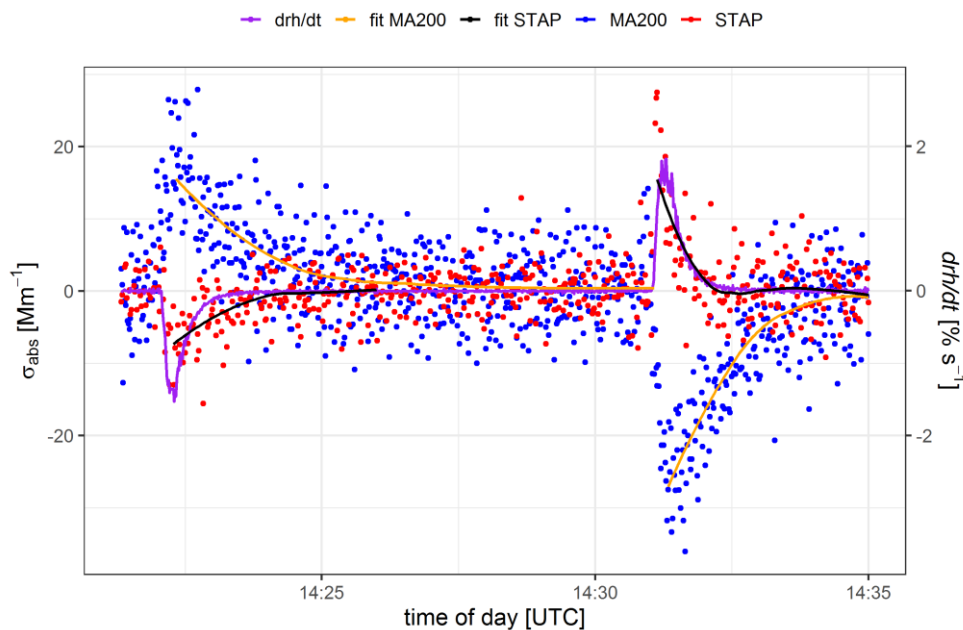
Key point 10: "*L325-326. Please show a scatter plot of the exponential decay, not only the time series.*"

Figure 1: 1 Hz raw data of σ_{abs} at 625 nm measured by the MA200 (blue points) and STAP200 (red points), the smooth fit through the measurements (orange and black) and d*rh*/d*t* (purple line)."

Key point 11: "*Explain clearly in the text and in the figure captions what is the difference between figs 3 and 5 and figs 4 and 6.*"

Response:

Thanks for that comment. Caption of Figure 4 is now: "Figure 4: Time series of *rh* (top panel) and absorption coefficient (bottom panel) measured with STAP (624 nm; black) and MA200 (625 nm; red) with clean filters." and of Figure 6: "Figure 6: *rh* of the air stream sampled by the MA200 and the STAP (upper panel) and σ_{abs} measured by MA200 and STAP at 625 (624) nm (lower panel). First up and down ramp of *rh* conducted with clean filter, second and third under conditions with filter loaded with ammonium sulfate. Loading periods around 18:30 and 21:00 UTC.". Caption of Figure 5 states now: " Figure 5: Scatter plot (dots) of all observations of the absolute excursion of σ_{abs} ($\Delta\sigma_{abs}$) in dependence of the absolute change in *rh* (Δrh), its linear regression fit as well as the summarizing boxplot of the linear regression fit are shown for the three investigated states (clean, loaded with BC and ammonium sulfate) at 624 nm (STAP, black colors) and 625 nm (MA200, red colors). Descriptive coefficients are given in Appendixtable 1." and correspondingly shows Fig. 7 the same plot but only considers the maximum and minimum areal loading density of the respective loading material. Caption of Fig. 7 states now: " Figure 7: Scatter plot of change in absorption ($\Delta\sigma_{abs}$) in dependence of the absolute change in *rh* (Δrh) separated into the different loading states (loaded with BC and ammonium sulfate) and minimum and maximum loading areal density on the filter. Dashed and colored lines represent the linear regression fit. Red and blue colors indicate MA200 at 625 nm and black and green colors indicate STAP at 624 nm. In the first panel BC loading is shown whereas in the second panel the ammonium sulfate case is displayed. Coefficients of the linear regression fit are displayed in panel 3. Shading of color in the linear fits and of the points are same as in panel 3."

The main difference between Fig. 4 and Fig. 6 is that in Fig. 6 smaller, stepwise *rh* changes were conducted. We updated the paragraph 3 in Section 3.2.2 to: "Figure 6 shows exemplarily the time series of *rh* of the sampled air *rh* and of the σ_{abs} measured with STAP and MA200 operated with clean filters. A *rh* of 0.0 to 96.2% with *drh/dt* humidity change rates of in the range of -1.42 and to 1.09 % s^{-1} was measured. Compared to the case in Figure 3 here a step-wise change of *rh* is shown. These steps resulted in a smaller absolute excursion of σ_{abs} which ranges from -7.2 to 9.0 Mm^{-1} (STAP; 624 nm, 60 s measurement resolution) and -14.1 to 10.9 Mm^{-1} (MA200; 625 nm, 60 second running mean). Furthermore, Figure 6 shows the response of the σ_{abs} to *rh* changes at three different states of filter loading. During the first ramp the filter were clean, during the second period the filters had a filter areal loading density of 32.5 (STAP) and 12.4 (MA200) mg m^{-2} and during the third ramp the filter in the STAP had loading areal density of 98.7 mg m^{-2} and the MA200 filter was loaded with an areal loading density of 37.6 mg m^{-2}. The response of the instruments during these periods is shown in Appendixtable 1."

The difference between Figure 4 and Figure 6 is that Fig. 4 shows the overall (mean) behavior of both instruments in BC and Ammonium sulfate case whereas Fig. 6 shows the response behavior of both instruments in cases of minimum and maximum loading of BC and Ammonium sulfate. The first sentence in paragraph 4 Section 3.2.2 already states now: "In **Fehler! Verweisquelle konnte nicht gefunden werden.** (lower left panel), the overall (mean) response of both instruments to *rh* changes is shown in the case of loading with ammonium sulfate." The second last sentence in the last paragraph of Section 3.2.2 already stated: "In **Fehler! Verweisquelle konnte nicht gefunden werden.** (middle panel), the spread of the slopes within the shown cases is exemplarily shown for the investigated minimum and maximum load of the filters.".