

# Response to review – RC3

## 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 #1

Remark: „Düesing et al. have provided a systematic and detailed characterization of the bias of two widely-used commercial absorption photometers which results from exposure to step RH changes. While they have not solved the problems of these photometers, they have nevertheless provided useful quantitative data and useful correction schemes. The manuscript should be published in ACP after addressing all of my comments below. The most important comments are that the data must be weighted by uncertainties before fitting, that the running mean the authors used has smoothed the data (and likely biased the fits), and that complete uncertainties must be provided for the authors' correction schemes.”

## Response:

We thank for the review. We will consider the general remark. Regarding the weighting of the fittings. The *rh* sensor uncertainty is constant in absolute value of 1.8% up to 90% *rh*. But the sensor includes also a response time which has to be considered as well. This is a very complex task and we cannot estimate the uncertainty induced by that. Therefore a weighting by uncertainties is not possible. The reviewer provided detailed comments and ongoing we will address them point by point:

Key point 1: “Section 2.1 (Theory of instruments) should be expanded to include mathematical statements of how the authors view the transient RH effects. In particular, it should be spelled out that  $M_{eBC}$  is based on the difference between subsequent attenuation measurements. This differential attenuation measurement also raises the possibility of investigating and correcting RH effects by looking directly at attenuation data. The authors should either look into this possibility, or discuss why they did not.”

## Response:

Thanks for that comment. We did not consider the attenuation because for most of the users of this instruments the particle light absorption coefficient and the eBC mass concentration are more intuitive.

Considering the effect of water in form of water vapor expressed as relative humidity we changed after Eq. (5): “ *Water has a refractive index of  $1.33+i1.5e-9$  at 532 nm wavelength. Hence it interacts with incoming electromagnetic radiation. If the filter is exposed to a relative humidity changes the light attenuation of the filter changes simultaneously, since the water binds to the filter itself (Caroll, 1976 and Caroll, 1986). Since a variety of filter materials, with different physical properties exist, we suspect that magnitude and sign of the light attenuation coefficient can vary with the filter material. The hypothesis is that the change rate of the *rh* ( $drh/dt$ ) directly determines the magnitude of the particle light absorption*

*coefficient, which depends on the difference of two subsequent attenuation measurements.”. We think that clarifies the paragraph.*

Key point 2: *“The use of a running mean for the MA200 means that the results are not equivalent to the 1-minute mean of the STAP. The running mean approach needs to be reconsidered. First, a running 60-second mean results in smoothing since each data point is used 3 times. Therefore the linear fits and R2 values reported are invalid since R2 is artificially enhanced by the autocorrelation which is inherent in a running mean. Best practice would be to analyze the 1-sec MA200 data and 60-sec mean MA200 data. The difference will provide insight into the STAP’s limited time resolution. This point is related to my next point.”*

and

Key point 3: *“The changes in Figure 3 are rapid relative to the 1-minute averaging intervals used. This means that the signal cannot be accurately represented by a single value (mean) during periods of change (increasing/decreasing RH). I would predict that increasing/decreasing RH periods have systematically different biases in the residuals of Figure 4. To correctly account for these biases, uncertainties must be estimated and an orthogonal regression must be performed in Figure 4, after weighting by these uncertainties. Most scientific software packages support this. Afterwards please highlight periods of increasing/decreasing RH in Figure 4 (eg with different symbols).”*

### Response to point 2 and 3:

Thanks for that comments. We choose the given averaging periods to show the general behavior of both instruments. For correction, the 1 Hz data of instruments were used. The linear regression is not artificially enhanced since we selected discrete points in the time series for the correlation. We described the method in the beginning of Section 3: *“This chapter will give an overview of the measurement results. The overall behavior of both instruments will be shown for wavelengths of 624 nm in the case of the STAP and 625 nm in the case of the MA200, respectively. A closer look at the behavior of both devices at 1 Hz time resolution shows that both devices differ greatly in quality. The STAP reacts very fast to relative humidity changes (see Figure 3, red dots and orange line) and then returns relatively fast to the zero line. The MA200, on the other hand, also shows a fast response to relative humidity changes, but then shows a distinct exponential recovery (see Figure 3) and reports absorption coefficients different from zero although there is no rh change.*

*Therefore, we use an averaging on a 60 second basis to describe the qualitative behavior of both devices. In the case of the STAP, the internal 60 second averaging is used. For the MA200, on the other hand, a 60-second “running average” is applied to the 1 Hz measurements.*

*The qualitative behavior of both devices is shown as follows. To each absolute change in the relative humidity ( $\Delta rh$ ) the corresponding maximum of the excursion of the averaged absorption coefficient ( $\Delta \sigma_{abs}$ ) has been assigned. Where the absolute change of the relative humidity is the difference between the relative humidity at the time of the largest excursion in the absorption coefficient and the relative humidity at the start of the excursion. This approach also excludes the response time of the rh sensor.*

*First, the results for the pure filter effect will be shown. Afterwards, we present the results of the combined behavior of filter and aerosol particles on the filters. For loaded filters, the combined effect will be shown separated into BC and ammonium sulfate loaded filter.”*

Furthermore, the STAP has no limited time resolution since it reports the raw intensities with a 1Hz resolution and from which the light absorption coefficient can be calculated based on Eq. (5). Anyhow, the different behavior of both instruments on a 1 Hz measurement resolution (exponential recovery of the MA200, see Fig. 3) led to the conclusion that we apply a running mean to the data of the MA200, which is similar to the internal running average of the STAP to describe the qualitative behavior of both instruments on that time base. Also, we included, that the *rh* sensor has a response time ( $t_{63}$ ) of less than 10 s (see sensor specifications) as the 2<sup>nd</sup> last sentence in paragraph 3 of Sect. 2.3: “*Furthermore, this sensor has a response time  $t_{63}$  of <10s.*”. The correlation of discrete points of the time series also accounts for that response time.

The scatter plot in Fig. 4 (now Fig. 5) shows the absolute change of absorption in dependency of the absolute change of *rh*. Therefore, the last non-excursed value (before *rh* change) of the absorption coefficient was subtracted from the maximum value of the absorption coefficient during the excursion. This was assigned to the difference of the starting *rh* and the *rh* at the point of maximum excursion of  $\sigma_{abs}$ . To clarify we included the description of the method as shown above. We furthermore think, that it was unclear that we did not correlate the whole time series of *rh* and  $\sigma_{abs}$  but the difference of discrete points in the time series. We think the updates make this clearer. Therefore, the points in Fig. 4 (now Fig. 5) do not need another symbol since points estimated based on a positive *rh* change are located in the right half of the scatter plots.

Key point 4: “*The authors have speculated extensively about the cause of the opposite trends of quartz and PTFE (lines 197-200). This speculation is of little value without experimental support. But I am not requesting experimental support. I am rather suggesting that the authors use these insights to design an improvement — use a mixture of the MA200 and TAP approaches to cancel out some of the biases of each approach. The utility of this suggestion can be tested by “simulating” a new instrument using the authors’ measurements. The design details related to feasibility of this should be commented on.*”

## Response:

Thanks for that comment. Since the underlying processes, especially the exponential recovery behavior of the MA200, are not fully understood. A new instrument could contain two sampling spots with both filters to cancel out each other. For that the different magnitudes of the effects have to be considered as well. It is only speculative why the PTFE filter inside the MA200 shows a larger response to *rh* changes. Besides the flow not passing through the reference spot, also the lower flow rate could have an effect but was not investigated. Therefore, we think a “simulation” of a new instrument is not useful right now. Anyhow, we included a paragraph, which gives a first idea how such a new instrument could be designed: “*Since the filter in the STAP reveals a positive and the filter in the MA200 a negative correlation to relative humidity changes a combination of both filters within one instrument could account for the observed effect. A new developed instrument could use these two different filter materials on two sampling spots to cancel out the effect of each other. Though, more investigations have to be done, especially to understand the different recovery behaviors and effect magnitudes of the PTFE and quartz-fiber filter.*”.

Key point 5: *"In Figure 7, why did the authors not simply sample for a longer time with the MA200 in order to match the loadings on either instrument?"*

**Response:**

Thanks for that comment. The loading of the STAP and MA200 under the given loading conditions can be converted to equivalent sampling periods of several hours depending on the prevalent ambient aerosol mass concentration. We wanted to keep these equivalent sampling equal for both instruments. Furthermore, under real life conditions the MA200 samples less particulate matter than the STAP by default due to the smaller flow rates.

Key point 6: *"Line 260, not only the imaginary part of refractive index but also the real part will affect these results, since the real part will influence scattering (influencing attenuation as well as subsequent absorption). Please reword."*

**Response:**

Thanks for that comment. We overthought this and came to the conclusion that the imaginary part does not have an impact on the shown behavior since it is too small in the case of ammonium sulfate. We reworded also following the comments of anonymous referee #4 and we updated the 5<sup>th</sup> paragraph in Section 3.2.2 to: "As shown in Fig. 5, both absorption photometers measure an "apparent" absorption coefficient of approximately  $2 \text{ Mm}^{-1}$  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 ( $0.15 \text{ Mm}^{-1} \%^{-1}$  for a clean filter,  $0.21 \text{ Mm}^{-1} \%^{-1}$  for ammonium sulfate, and  $0.30 \text{ Mm}^{-1} \%^{-1}$  for BC). Ammonium sulfate has a real part of  $1.521 \pm 0.002$  (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 cause 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 8: “The correction schemes are not perfect, but they are useful. Certainly these and other authors will apply them at some point. It is therefore very important to report UNCERTAINTIES for the correction schemes. Both a percentage uncertainty and a bias (absolute value, in analogy to limit of detection) uncertainty must be reported. The bias requirement is illustrated in Figure 10, where 2/Mm of false signal result from a step RH change of about 30%. This bias of 2/Mm means that a true signal of 1/Mm would hardly be measurable.

I do not know of a formal reference for handling this kind of bias, but I have encountered it in my own work and thought a bit about an easily understandable solution. My best suggestion is to allow users to answer the question: what is the minimum reported value which I can trust, if I am willing to accept a maximum inaccuracy of 25%? This question can be answered with a simple mathematical formulation which I will leave for the authors to provide. The answer to this question (the actual bias) will obviously depend on the magnitude of  $\Delta rh$ .”

## Response:

Thanks for that comment. We reworded a major fraction of Section 3.3.1 including the correction scheme of the STAP. It states now:” In **Fehler! Verweisquelle konnte nicht gefunden werden.** the correlation of  $rh$  change rate ( $drh/dt$ ) and the measured  $\sigma_{abs}$  at 624 nm measured by the STAP (red circles) and recalculated with respect to standard conditions (pressure of 1013.25 hPa and temperature 273.15 K) is shown. The STAP-based background eBC mass concentration during the experiment was  $\sim 190 \text{ ng m}^{-3}$  (at standard conditions,  $\sigma_{abs}$  at 624 nm converted with a MAC of  $6.6 \text{ m}^2 \text{ g}^{-1}$ ), which corresponds to offset (standard conditions corrected values) in the shown scatterplot of **Fehler! Verweisquelle konnte nicht gefunden werden.** and which has no influence on the response to  $rh$  changes as shown previously.

The  $rh$  change rate ranged from  $-10.8$  to  $14.5 \text{ \% s}^{-1}$ . These rates correspond to a  $\sigma_{abs}$  of  $-231$  to  $192 \text{ Mm}^{-1}$  for recalculated values at standard conditions and  $-203$  to  $164 \text{ Mm}^{-1}$  directly measured by the instrument. But these measurements are biased by the response time of the relative humidity sensor so that the “real”  $rh$  change-rate cannot fully represented by these measurements. On average the slope (correction factor  $C_{rh}$  in Eq. (8)) of the linear fit is  $10.08 (\pm 0.12) \text{ Mm}^{-1} \text{ s \%}^{-1}$  for standard conditions and  $8.82 (\pm 0.10) \text{ Mm}^{-1} \text{ s \%}^{-1}$  for direct instrument output, respectively. Calculating the particle light absorption coefficient introduced by  $rh$  changes with:

$$\sigma_{abs,rh} = C_{rh} \frac{drh}{dt} \quad (8)$$

for different  $rh$  change rates in both, the recalculated and direct instrument output case, and subtracting it from measurements allows to correct for the observed effect as follows:

$$\sigma_{abs,corr} = \sigma_{abs,meas} - \sigma_{abs,rh} , \quad (9)$$

and after replacing  $\sigma_{abs,rh}$  in Eq. (8) with Eq. (9) follows:

$$\sigma_{abs,corr} = \sigma_{abs,meas} - C_{rh} \frac{drh}{dt} . \quad (10)$$

The y-intersect of the linear fit in **Fehler! Verweisquelle konnte nicht gefunden werden.** has not to be considered for correction as mentioned before. Disadvantageously, with this correction the noise of the

*rh* sensor will propagate in the corrected  $\sigma_{\text{abs}}$ . Furthermore, the linear fit in **Fehler! Verweisquelle konnte nicht gefunden werden.** under- or overestimates the behavior in regimes of very high relative humidity change rates most likely due to the response time of the *rh* sensor, so that the correction function cannot entirely correct the bias. Therefore, the given correction factor  $C_{\text{rh}}$  consists of uncertainties, which cannot be entirely addressed. Hence, it is only a first guess, needs further refinement and right now we do not recommend to use the correction approach as long the uncertainties are not fully addressed. Furthermore, since only one STAP was tested, other STAP may have other correction factors due to a unit to unit variability. Additionally, other filter materials used in the STAP can also lead to another behavior. Anyhow, the upper function was applied to STAP measurements conducted with the same *rh* sensor under atmospheric conditions.

Exemplarily, **Fehler! Verweisquelle konnte nicht gefunden werden.** shows this application. The figure shows airborne measurements of  $\sigma_{\text{abs}}$  at 624 nm derived with the STAP derived during a campaign conducted in March 2017 in East Germany. The upper panel displays the *rh* of a dried aerosol sample stream measured upstream of the STAP. The lower panel shows the recalculated  $\sigma_{\text{abs}}$  at 624 nm wavelength corrected for *rh* changes (black) and biased by *rh* changes (red). In the periods where the *rh* changes relatively fast ( $drh/dt$  of -0.55 to 0.56 % s<sup>-1</sup> e.g. at around 6200 seconds), the uncorrected  $\sigma_{\text{abs}}$  overshoots. The correction significantly reduces this bias and smooth out the measurements during the periods of *rh* changes. At the peaks of  $drh/dt$  the difference of the corrected and uncorrected values is up to 1.5 Mm<sup>-1</sup>, which is significant with respect to the measured  $\sigma_{\text{abs}}$ . The periods with negative  $\sigma_{\text{abs}}$  are not introduced by the *rh* effect. We moreover think that a small offset is introduced in the initialization process of the instrument. Despite the imperfection of the correction scheme, this linear approach can be useful to derive a rough estimate of the accuracy of the measurements. For instance let  $x$  be the required accuracy for the measurements in % and  $\sigma_{\text{abs}}$  the measured particle light absorption coefficient we can express the ambient particle light absorption coefficient which is at least needed to fulfill the accuracy criterion in dependency of the *rh* change rate  $drh/dt$ :

$$\sigma_{\text{abs, meas}} \geq \frac{100\%}{x[\%]} C_{\text{rh}} \left[ \frac{\text{Mm}^{-1}\text{s}}{\%} \right] \left| \frac{drh}{dt} \right| \left[ \frac{\%}{\text{s}} \right]. \quad (11)$$

Exemplarily, if a change rate of 0.1 % s<sup>-1</sup> is measured and an accuracy of 25% is needed, at least a measured particle light absorption coefficient of around 4 Mm<sup>-1</sup> is needed to fulfill the accuracy criterion.”.

The main reason why we did not provide any uncertainties of the correction scheme besides the uncertainty of the slope is that we simply cannot quantify the uncertainty introduced by the response time of the *rh* sensor. Furthermore, each *rh* sensor will have different characteristics so that the correction scheme, if any, can only be applied using this sensor. We moreover suggest to use the findings to estimate the measurement uncertainties introduced by *rh* changes and to set a lower threshold of reliable measurements depending on the required accuracy and prevalent *rh* change rate.

For the MA200, the problem is even more complex since the correction approach results in slightly different coefficients of the correction formula when applied to other similar experiments. This could be due to a unit-to-unit variability or other phenomena affecting the PTFE response so that not all uncertainties can be addressed. Furthermore, the response time of the *rh* sensor introduces some uncertainty to the correction approach which cannot be quantified. Since, we do not recommend to use the correction scheme providing uncertainties is of little value. Similar to the STAP the correction approach looks a) promising and shows the right direction and b) could be used to roughly estimate the bias in the

measurements due to *rh* changes. Also, the last sentence of the second last paragraph and the last paragraph of Section 3.3.2 states now: *"Here, the response time of the sensor could account at least for a part of the imperfection of the correction approach and cannot be fully quantified, yet."*

*Unfortunately, the application of the same correction approach to other similar experiments resulted in different correction function  $a$  and  $b$ . Applying the approach to two clean case experiments from section 3.1 resulted in optimized parameters of  $a = -0.92$  and  $-1.03$  and  $b = 0.974$  and  $0.971$ , respectively. Hence, it is just a first step trying to account for relative humidity changes and further research with more MA200 simultaneously has to be done to fully understand the underlying processes and to fully quantify the uncertainties of the correction scheme. Nevertheless, the presented approach significantly reduces the amplitude of the bias in the shown data set (see **Fehler! Verweisquelle konnte nicht gefunden werden.** 10). But, up to now we cannot recommend to use the given parameters to correct for *rh* effects. At most it can be used to make a rough estimate of how measurements of the particle light absorption coefficient derived the MA200 could be biased by *rh* changes."*

#### Minor comments:

We thank the reviewer for all the minor points, we will comment on each separately point by point.

Point 1: *"1. I would suggest taking the natural logarithm of Equations 1 and 2, or at least 2, so that the important terms (exponents of  $e$ ) are more easily visible. Also, please at line 99 add a sentence clarifying that reinterpreting  $l$  as an aerosol path length does not mean that  $\sigma$  represents the aerosol absorption coefficient but still the filter attenuation coefficient."*

#### Response:

We expressed Eq. (1) and Eq. (2) following the reviewer. In principle the theory should explain how the particle light absorption coefficient is derived. We followed the recommendations of the reviewer and added the sentence: *"But, a reinterpretation of the path length does not mean that the result is the particle light absorption coefficient, but still the light attenuation coefficient."* But, we used the term light attenuation coefficient instead of filter attenuation coefficient.

Point 2: *"Line 116, please change "provide" to "report" since the photometers only estimate eBC."*

#### Response:

We changed according the referees comment.

Point 3: *"Comparison" by who, are those unpublished results from the authors' lab?*

#### Response:

Yes, these are unpublished results from the authors' lab and the data can be requested if needed. We changed the last paragraph in Sect. 2.1 to: *"Lab-comparison of the eBC mass concentration between a MAAP (Multi Angle Absorption Photometer; Thermo Fisher Scientific, 27 Forge Parkway, 02038 Franklin,*



MA, USA; Petzold and Schönlinner, 2004) at 637 nm wavelength and MA200 at 625 nm and STAP at 624 nm *beforehand the experiment* revealed a good agreement within 3% and within 6%, respectively. For the STAP a MAC of  $6.6 \text{ m}^2 \text{ g}^{-1}$  was assumed. Since a MAC of  $6.6 \text{ m}^2 \text{ g}^{-1}$  is used for the MAAP at 637 nm, in this study we used the  $\sigma_{\text{abs}}$  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.”

Point 4: “Line 138 and 155, I suggest SI units of area”.

#### Response:

Thanks for the comment. We are not sure what the reviewer means with Si-units of area. To our opinion  $\text{m}^2$  is already a SI-unit.

Point 5:” Line 167, change “by passing” to “by passing it through” (this sentence required 3 reads to be understood)”

#### Response:

We updated to: “One of the flows was humidified by passing *through* two glass tubes containing distilled water at room temperature with an inlet and outlet for compressed particle free air.”.

Point 6: “I have not seen the term “floating mean” used before and an internet search did not bring up any definitions. I would recommend “running mean” (more precise, since floating implies complete freedom whereas running implies autocorrelation).”

#### Response:

We changed each occurrence of floating mean with running mean.

Point 7: The Section “Recommendations” should be a numbered section or subsection, and no sections should come after Conclusions.

and

Point 8: In Recommendations and the Introduction, the authors suggest avoiding fast changes by ascending slowly. This is simply not possible in some scenarios (unmixed layers, clouds) and this should be noted.

#### Response to point 7 and 8:

Thanks for the comments. We removed the heading of the recommendations section and changed to the paragraph with the recommendations to: “The findings summarized above lead to following recommendations how to use this type of instruments:

1. When used for vertical profiling, apparent sharp gradients in *rh* during the profile have to be taken into account.



- a. The ascending speed of the profiling platform should be reduced if possible, to decrease the temporal change of  $rh$ , but in some scenarios this is simply not possible and therefore,
  - b. when fast relative humidity changes cannot be avoided, such periods have to be removed from the data set, or at least to estimate the uncertainties of the measurements based on the presented correction functions. Therefore,
2. we recommend recording the  $rh$  of the sampled aerosol. This allows to determine  $rh$  change rates. This allows to roughly estimate the bias of  $rh$  changes on filter-based absorption measurements with these two instruments.
  3. The usage of a dryer is highly recommended, because it reduces the amplitude of the excursion in the measurements during fast  $rh$  changes.
  4. For both instruments we recommend to conduct more similar experiments to address the flaws of our study to refine the presented correction approaches.
  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.”.

Also, we added in the last paragraph of the Abstract: “Due to our findings, we recommend to use an aerosol dryer upstream of absorption photometers to reduce the  $rh$  effect significantly. Furthermore, when absorption photometers are used in vertical measurements, the ascending or descending speed through layers of large  $rh$  gradients has to be low to minimize the observed  $rh$  effect. **But this is simply not possible in some scenarios especially in unmixed layers or clouds.** Additionally, recording the  $rh$  of the sample stream allows correcting for the bias during post processing of the data. This data correction leads to reasonable results, according the given example in this study.”.

Point 9: “Table 1: I see no bold entries.” and point 10: “Table 2: Instead of custom formatting, add a column “Filter Number” which increases by 1 when appropriate.”

**Response:**

Thanks for the comment. We updated the tables to:“

**Table 1:** Filter loading mass concentration ( $M_{eBC}$ ) of the black carbon particles and filter areal loading density (deposited mass per spot area)  $\rho^*_{eBC,i}$ .  $M_{eBC}$  were determined by dividing the average  $\sigma_{abs}$  of the STAP with an assumed MAC of  $6.6 \text{ m}^2 \text{ g}^{-1}$  or based on the MAAP measurements. Usage of same filter is indicated by its filter number. Bold written entries were used for the investigation of the  $rh$  effect.

filter number	$M_{eBC}$ [ $\mu\text{g m}^{-3}$ ]	$\rho^*_{eBC,i}$ [ $\text{mg m}^{-2}$ ]	
		STAP	MA200
#1	<b>44.5 (STAP)</b>	<b>14.0</b>	<b>5.4</b>
	43.4 (STAP)	37.9	14.4
	<b>27.6 (STAP)</b>	<b>42.9</b>	<b>16.3</b>
#2	<b>52.6 (MAAP, 2 scans)</b>	<b>2.8</b>	<b>1.1</b>
#3	-	<b>13.7 (integral of STAP)</b>	no data

**Table 2: Average volume and mass concentration ( $V_{(\text{NH}_4)_2\text{SO}_4}$ ,  $M_{(\text{NH}_4)_2\text{SO}_4}$ ) of the loading  $(\text{NH}_4)_2\text{SO}_4$  aerosol derived from the used MPSS (number of used scans in brackets) and loading areal density  $\rho^*_{(\text{NH}_4)_2\text{SO}_4}$  of the filters are given. Usage of same filter is indicated by its filter number, which means that the filter loading mass was adding up during the experiments.**

filter number	$V_{(\text{NH}_4)_2\text{SO}_4}$ [ $\mu\text{m}^3 \text{ cm}^{-3}$ ] (# scans)	$M_{(\text{NH}_4)_2\text{SO}_4}$ [ $\mu\text{g m}^{-3}$ ]	$\rho^*_{(\text{NH}_4)_2\text{SO}_4}$ [ $\text{mg m}^{-2}$ ]	
			STAP	MA200
#1	15.4 (2)	27.2	3.1	1.2
	18.6 (1)	32.9	10.5	4.0
	20.6 (3)	36.4	31.3	11.9
#2	20.6 (4)	36.5	40.8	15.5
#3	33.1 (3)	58.6	32.5	12.4
	33.5 (5)	59.3	98.7	37.6
#4	20.3 (3)	36.0	21.1	8.0
	20.3 (3)	36.0	41.9	15.9
#5	23.9 (3)	42.4	28.9	no data
	28.4 (4)	50.2	69.8	no data
	29.8 (2)	52.8	99.6	no data