

## Response to Reviews

Manuscript amt-2014-257

### The “Dual-Spot” Aethalometer: an improved measurement of Aerosol Black Carbon with real-time loading compensation

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We thank the referees for their comments. We have revised the manuscript to incorporate the feedback to all comments and advice. We have copied the remarks of each Referee in *black italics* and our responses are given in regular black font. Manuscript text with revisions is given in [regular blue font](#).

Some of the comments given by the different referees seem to be in conflict, especially regarding the supplemental information. We have considered the instructions in a way, which maximizes the information conveyed to the reader.

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#### Anonymous Referee #1

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#### **General**

*The aethalometer has been used for measuring black carbon concentrations for decades without any significant development of the instrument even though it has been observed that there are some inherent problems related to the method in general. The company has obviously taken seriously the observations made by several scientists in the field and developed the method in a very interesting and promising way. This paper presents the modification and how the new version compares with other instruments. The results seem good and the instrument even presents a way of obtaining new and interesting kind of information on the optical properties of aerosol. The paper is valuable and definitely worth publishing in AMT. However, in my opinion there are some points (see below in detailed comments) that should be explained in more detail, otherwise they lead to endless speculations and discussions within the community.*

We thank the referee for her/his comments and address the points she/he raised in more detail below.

## Detailed comments

**P10183 L15-16** “loading effect differs between the seasons, possibly depending on the aerosol properties”.

*I wonder why there is the word “possibly”? What else could the loading effect even hypothetically depend on but aerosol properties? If an aerosol measurement instrument works properly, and measurement conditions are kept stable, only aerosol affects the result, all other disturbances are errors.*

The Referee is correct and we have modified the manuscript accordingly, citing an additional reference. The main text has been modified as follows (page P10183, L14-16) :

“However, it has been shown that the loading effect differs between the seasons, depending on the aerosol properties (Virkkula et al., 2007). Weingartner et al. (2003) have shown that fresh aerosols exhibit more pronounced loading effects than aged ones.”

**P10188, L8.** “Gundel et al. (1984) showed that optical attenuation measured at 532 nm ... “. I downloaded the Gundel et al. article. I did not find anywhere the wavelength they used. In their section “Attenuation measurements”, p 198 of the paper, it is written only “The laser transmission method measures the attenuation of visible light as it passes through the filter.” So, do the authors have some additional information on the wavelength or should they refer to some other paper?

We thank the Referee to point out this error – the laser used in the experiments at the UC Lawrence Berkeley Laboratory was in fact a He-Ne laser (633 nm) even though the wavelength or the laser type are not mentioned in the Gundel et al. article. The attenuation measuring apparatus is explained in Rosen et al. (1978) which is referenced in Gundel et al. (1984). We hence amended the text in the manuscript to (page P10188, L8) and added the Rosen et al., 1978, reference:

“Gundel et al. (1984) showed that optical attenuation measured at 633 nm (Rosen et al., 1978) for samples collected on quartz filter started to deviate from the linear relationship as a function of surface loading (B) above 10  $\mu\text{g cm}^{-2}$  of black carbon.”

**P10188,** The authors give a parameterization of ATN vs. loading B as

$$ATN = \frac{1}{k} (1 - e^{-kB\sigma}), \quad \text{Eq. 4}$$

*This is very different from that of Gundel et al. (1984) who give this parameterization in their eq. (5):*

$$ATN = -100 \ln(\alpha + (1 - \alpha)e^{-\sigma B})$$

I plotted both of these in the same figure, ATN vs B, Fig. 1 below. For the present paper's eq. (4) I used the values  $\sigma = 13 \text{ m}^2 \text{ g}^{-1}$  and a few  $k$  values and for Gundel et al., eq. (5) their values  $\alpha=0.0172$ ,  $\sigma = 23.9 \text{ m}^2 \text{ g}^{-1}$ . With the present paper's eq. (4) I could not get the same shape and values as Gundel et al with any  $k$  &  $\sigma$  combination. There is probably an infinite number of ways to make a curve that saturates at some level. So, how did you arrive at Eq. (4)? Has it only been chosen simply to get some way to derive Eq. (6)?

There are indeed a number of functions, which can be chosen to show saturation at higher values of the function parameter, the  $(1 - e^{-t})$  function is a well-known example. We explicitly mention that this is an empirical relationship (page 10188, L10):

“This can be parameterized by an empirical relationship of ATN vs. loading B:...”

It is also different from the one in Gundel et al. (1984), because there the assumption was that there is a fraction of light being guided within the fibers and “bypassing” the particles collected on the filter fibers, causing the saturation; we now know that the mechanism is much more complex. We have observed a linear relationship between the non-compensated BC and ATN (Fig. 2), so had others (Park et al., 2010), hence we have chosen the saturation function giving a linear-like relationship of BC as a function of ATN.

**P10188 L24-25** “This equation is similar (but not the same) to the one presented by Virkkula et al. (2007)”. I started wondering, do these equations have something in common. They do.

The Drinovec et al. Eq. (6) is  $BC = BC_{NC} \frac{1}{(1-k*ATN)}$

On the other hand, the sum of a geometric series

$$\sum_{n=0}^{\infty} r^n = 1 + r + r^2 + r^3 + \dots = \frac{1}{1-r} \text{ for } |r| < 1$$

If  $|kATN| < 1$ , eq. (6) can be written

$$BC = BC_{NC} (1 + kATN + (kATN)^2 + \dots)$$

For small values of  $|kATN|$  the terms  $(kATN)^n$  with  $n > 1$  are negligible so the first two terms are actually just the same as in Virkkula et al. (2007). This could be mentioned in the paper so that those that have already determined compensation parameters from the old aethalometer data know it is not fundamentally different from the new one – except that for different filter material it is somewhat different.

Writing Eq. (6) as a geometric series is indeed very instructional: the first two terms in the expansion of the series are in fact identical to the Virkkula et al. (2007) compensation scheme, the two approaches are in fact equivalent to the first order (that is, neglecting the higher order terms). However, different filter material needs to be taken into account when comparing the compensation parameter values for the AE31 and AE33 (see sections 3.2 and 3.3 and the

associated Figures 5, 6 and 7). We have included this comparison in the text, expanding the paragraph on the different compensation methods (page P10188, L24-25):

“This equation is similar (but not the same) to the one presented by Virkkula et al. (2007) (Table 1). In fact, expanding Eq. (6) gives a geometric series, which, in the zero and first order, gives the same dependence of the non-compensated black carbon concentration  $BC_{NC}$  on ATN as the one described in by Virkkula et al. (2007).”

**P10189, L15.** *Sensitivity of  $k$  to flow measurements. How sensitive is it and how accurate are the flow measurements? Give an uncertainty estimate for  $k$  with error propagation including uncertainties of flows and spot sizes.*

The accuracy of the flow measurement is determined by the accuracy of the flow-meter used for calibrating the instrumental flow. These are typically between 0.75% and 2%. Solving Eq. (9) at low ATN values is unstable. We add the face velocity ratio factor, determined at low ATN and hence independent of the loading effect to stabilize the numerical calculation. We have added a reference to the requested information in the manuscript text, immediately after Eq. (10):

“*FVRF* is determined from the  $ATN_2/ATN_1$  ratio obtained at low filter loadings. A comprehensive analysis on the sensitivity of the determination of  $k$  and the determination of *FVRF* can be found in the Supplement (Determination of face velocity ratio factor (*FVRF*), and Figs. S6 and S7).”

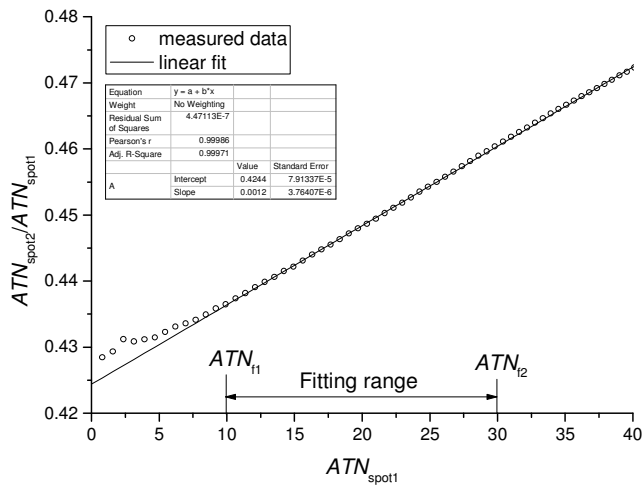
And an in-depth description to the Supplement:

#### “Determination of face velocity ratio factor (*FVRF*)

The loading effect compensation algorithm is sensitive to correct measurement of sample flow through the spots. Flow through the spot2 is especially susceptible to errors since it is calculated as a difference between total flow and flow through the spot1. *FVRF* is used to compensate for flow measurement uncertainty (Article, Equation 11):

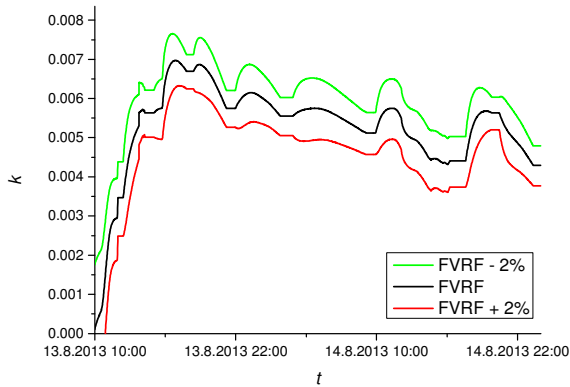
$$FVRF = \left( \frac{ATN_{spot2}}{ATN_{spot1}} \right)_0 \cdot \frac{F_1}{F_2}$$

The calculation of *FVRF* is based on the fact that at small filter loadings the attenuation is proportional to the flow through the spot. Because of the transients on the fresh spot, the first ATN measurements (where  $ATN_1 < ATN_{f1}$ ) are omitted from the analysis. Data with  $ATN_1$  (attenuation for channel1, spot1) between the lower limit  $ATN_{f1}$  and upper limit  $ATN_{f2}$  are used for determination of *FVRF* (Figure S6). The actual parameter which tells us the correct flow ratio is the intercept of the linear fit of  $ATN_{spot2}/ATN_{spot1}$  versus  $ATN_{spot1}$  (Figure S6). The average *FVRF* for channels 2 to 6 is used for further calculations.



**Figure S6.** An example of fitting  $ATN_{spot2}/ATN_{spot1}$  versus  $ATN_{spot1}$  data to obtain the intercept value. The intercept  $(ATN_{spot2}/ATN_{spot1})_{ATN=0}$  represents a real flow ratio  $F_{spot2}/F_{spot1}$ .

*FVRF* values differ slightly between the instruments and spots. Here is an example of mean values and standard deviation of *FVRF* for 5 different instruments:  $1.001 \pm 0.025$ ,  $1.059 \pm 0.011$ ,  $1.063 \pm 0.001$ ,  $1.017 \pm 0.006$  and  $0.979 \pm 0.02$ . If the flow ratio is not compensated, a wrong value of parameter *k* is obtained. The influence of 2 % change of flow ratio on the parameter *k* is presented in Fig. S7.



**Figure S7.** An illustration of the sensitivity of a dual spot loading effect compensation algorithm to a 2% change in the face velocity ratio factor (*FVRF*).

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**10189, L16-25.** *I don't really understand the way FVRF is determined. This is something that has to be explained in very detail, otherwise the community keeps analyzing and speculating this for ever. First, if flows are measured accurately, there should not be any such factor. If either one or both flow sensors have errors, their true values would be approximately  $F_{TRUE} = slope \times F_{AETH} + F_{OFFSET}$ . How does this lead to eq. (11)?*

*On L20 it is written "FVRF is determined from the  $ATN_2/ATN_1$  ratio obtained at low filter loadings".*

*The whole determination of  $k$  from eq. (9) is based on determining  $ATN_1$  and  $ATN_2$  – how can it then be used for determining flow errors? Why would it be different at low filter loadings?*

*On L 21-22 it is written "FVRF is determined as an intercept of the  $(ATN_2/ATN_1)$  vs.  $ATN_1$ ".*

*Intercept? You mean the value of the ratio when  $ATN_1 = 0$ ? That is infinite. This determination of FVRF needs to be explained so that even simple scientists like me understand it. Use perhaps graphical presentations.*

The parameter *FVRF* is dependent on the deposition of the sample on the filter. At low loadings of the filter, solving Eq. (9) proves unstable. At the same time, loading effects at low ATN are negligible. We use this fact to estimate the parameter *FVRF*, by fixing the value to the one determined by the fit in the ATN interval where loading effects do not influence the measurement. The intercept cannot become infinite because both  $ATN_1$  and  $ATN_2$  approach 0.

The detailed description on the determination of *FVRF* and the sensitivity of compensation has been added to the Supplement (please see answer just above).

**P10190,** *Calculation of  $k_{weighted}$ , eq. (12), is also very unclear. I understand calculating weighted averages but not this formula. Are there ATN values of both spot 1 ( $ATN_{TA}$  and  $ATN_1$ ) and spot 2 ( $ATN_{j2}$ )? Again, explain very clearly, derive the formula and possibly use also graphical presentation. Don't leave space for speculations and misunderstandings.*

We have unified the notations of ATN throughout the text, added a detailed description to the supplement to explain the smoothing and the calculation of the compensation parameter  $k_{weighted}$ . The supplement is referenced immediately before Eq. (12) in the main body of the manuscript:

*"For the compensation, we thus use a weighting method, where the previously-determined  $k$  from the fully loaded spot before the tape advance is taken into account (see also Supplement, Weighting method for loading effect compensation parameter  $k$ , and Fig. S8):"*

Supplement addition:

**“Weighting method for loading effect compensation parameter  $k$**

Determination of the parameter  $k$  at low filter loadings is very susceptible to small measurement errors. When more and more material is collected on the spot, the filter loading effect is easier to measure using the dual spot approach. To reduce the uncertainty of the parameter  $k$  at low attenuations, a weighting method is applied (main body of the article, Equation 12). Both weighted and un-weighted values of parameter  $k$  are presented on Figure S8.

For  $ATN_1 < ATN_{f2}$  a value of  $k_{old}$ , which is obtained from the previous filter spot, is reported:

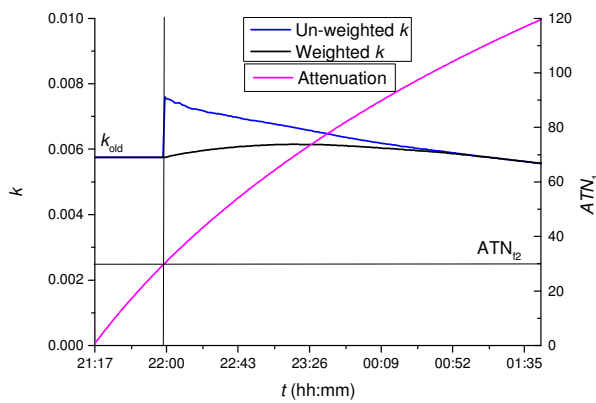
$$k_{weighted}(ATN_1 < ATN_{f2}) = k_{old}.$$

For  $ATN_1 > ATN_{f2}$  a weighted value of the parameter  $k$  is used, which takes into account the  $k_{old}$  and the un-weighted value of parameter  $k$ . The default value is  $ATN_{f2}=30$ . Progressively, as the attenuation increases, less and less of  $k_{old}$  is incorporated into  $k_{weighted}$ :

$$k_{weighted} = \frac{(ATN_{TA} - ATN_1)k_{old} + (ATN_1 - ATN_{f2})k}{(ATN_{TA} - ATN_{f2})}.$$

At the time of full spot loading, the weighted value of  $k$  becomes equal to the instantaneous (un-weighted) value:

$$k_{weighted}(ATN_1 = ATN_{TA}) = k.$$



**Figure S8. An example of the instantaneous and the weighted value of the parameter  $k$  during spot loading. For  $ATN_1 < ATN_{f2}$  the  $k$  value from the previous spot is used. For attenuations between  $ATN_{f2}=30$  and  $ATN_{TA}=120$ , a weighting method is applied.”**

**P10190 L14-15:** *“Because the air flow is measured after the air passes the filter, lateral air flow in the optical chamber has to be taken into account...”*

*There is nothing wrong here, I just want to congratulate the authors to have taken this into account, usually nobody thinks about this.*

Thank you. The sentence has been modified for clarity in response to referee #4’s comment:

*“Because the air flow is measured after the air passes the filter, lateral air flow in the filter matrix under the chamber  $\zeta$  has to be taken into account:” ...*

**P10191 L1-2:** *“The enhancement parameter  $C$  has no statistically significant spectral dependence (Weingartner et al., 2003; Segura et al., 2014),”*

*This is not true. Even some of the coauthors of the present paper are also coauthors in the latter paper, Segura et al. (2014) referenced in that sentence. There it is written for example this:*

*“To obtain the wavelength dependence of  $C(\lambda)$ ... ” and “... $C$  values obtained span from 3.42 at 370 nm to 4.59 at 950 nm, and their spectral differences ... ”. And there is other literature on this also. The  $C$  is in a way an artificial factor, it is not unambiguously derived from any first principles but it is generally considered to compensate for multiple-scattering effects in the filter-aerosol matrix. Scattering by aerosols depends on wavelength so also intuitively it is reasonable that  $C$  would be wavelength dependent.*

We agree, the parameter  $C$  is derived from the comparison of different measurement methods, each with its own issues and idiosyncrasies – there is no “reference” method or an ab-initio way to determine this parameter.

Deriving the parameter  $C$  from comparisons between different instruments introduces uncertainties and the experimental errors need to be taken into account. This relative nature of the parameter  $C$  is inherent to the procedure used to obtain it. We are working on a methodology to assess the filter based photometers’ (including Aethalometers) sensitivity to non-absorbing aerosols and an accurate determination of the parameter  $C$  using an “extinction minus scattering” technique to determine absorption which can be compared to absorption derived from filter based photometer measurements. We have reanalyzed the data reported for Granada (Segura et al., 2014) including the errors, uncertainties and/or variability in the measurements of absorption and attenuation by the MAAP and the Aethalometer AE31; the scattering coefficient by the TSI 3563 nephelometer; and the two associated Ångström exponents. The resulting variation in  $C$  is up to 0.8 in the infrared part of the spectrum (950 nm), the wavelength dependence of  $C$  being therefore possibly smaller than anticipated earlier. We have assessed the issue by examining the cross-sensitivity of the Aethalometers AE33 to scattering in a dedicated chamber experiment. We found cross-sensitivity, independent of the loading of the (see new section “Cross-sensitivity to scattering” of the manuscript, and the discussion to the question just below).



**P10191 L8-12:** “ BC is historically defined by Aethalometer measurement at 880 nm, so we use parameters at this wavelength to derive the mass equivalent concentration using  $\sigma_{\text{air}} = 7.7 \text{ m}^2 \text{ g}^{-1}$ , obtained initially by comparing optical and thermal measurements of filters loaded with refractory carbonaceous material (Gundel et al., 1984).”

*What is  $\sigma_{\text{air}}$ ? Why is the subscript “air”? Mass absorption coefficient of air? If it is MAC of BC, like I assume, where does the actual value  $7.7 \text{ m}^2 \text{ g}^{-1}$  come from? There is no number 7.7 and no data at the wavelength of 880 nm in Gundel et al. (1984), I have that paper in front of me. If it is MAC of BC, I still question the value. Bond et al. (2013) (JGR 118, doi:10.1002/jgrd.50171) write “Measured values for freshly generated BC fall within a relatively narrow range of  $7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$  at 550 nm.” If it is assumed that the MAC depends inversely on wavelength, it is much smaller at 880 nm.*

*At this point I jump to the supplement because the MAC table is presented there. How were the mass absorption cross sections in Table S2 obtained? In the supplement lines 97 – 89 it is written “The relationship between the absorption and mass was determined by optical measurements of transmission and thermal measurements of samples, where the nonrefractory constituents of the carbonaceous sample were removed (Gundel et al., 1984)”*

*This is definitely not detailed enough. This table will be a very important one in the future, all aethalometer users will refer to this table several times. So, it should be explained in detail. How did you produce the BC? What was the independent method for measuring BC or EC mass concentration. And so on.*

The parameter  $\sigma_{\text{air}}$  is indeed meant to represent MAC of aerosols freely suspended in the air, hence the subscript “air”. The enhancement of absorption in the filter matrix is described by a multiplication factor  $C$ . This assumes that the loading effects and the multiple-scattering in the filter matrix can be separated. We have shown with a laboratory experiment that loading of the filter matrix with varying amounts of scattering aerosols does not increase the apparent absorption (reported by the Aethalometers AE33) due to the accumulation of scattering particles on the filter: the parameter  $C$  does not depend on the amount of scattering particles on the filter (see section 3.7 below). Similarly, the size of the absorbing particles on the filter does not influence the parameter  $C$  (ACTRIS, 2014b). Both of these results are a subject of an ongoing effort for further characterization of filter based absorption photometers and a development of a reference method to determine aerosol absorption by the difference between extinction and scattering. We have added Section 3.7 describing these very important results, and scientists participating in the experiment were added as the authors of the manuscript.

### “3.7 Cross-sensitivity to scattering

The cross-sensitivity of the Aethalometer AE33 to the scattering particles collected in the filter matrix was tested in a laboratory experiment. Ammonium sulfate aerosols were used as model scattering aerosols. Ammonium sulfate was aerosolized in an atomizer from a solution. The

airstream was dried to relative humidity below 30% by mixing with dry clean air and additionally using diffusion driers. The particles were injected in the mixing chamber (volume 0.5 m<sup>3</sup>). Up to eight outlet ports with equal concentrations distributed the aerosol to the instrumentation. The scattering coefficient was measured with a polar nephelometer (Aurora 4000, Ecotech Pty Ltd). The truncation error was corrected using the methodology described in Müller et al., 2011b. The loading of the spot with ammonium sulfate was chosen to represent polluted atmospheres. Mass concentrations for the two experiments shown in Figure 12 were derived from the particle number size distributions and the density of 1.53 g/cm<sup>3</sup> for ammonium sulfate, and amount to 39 µg/m<sup>3</sup> and 43 µg/m<sup>3</sup>. The volume mean diameters were measured to be 156 nm and 257 nm, respectively.

The sensitivity of the determination of the absorption coefficient from the AE33 to the scattering is shown in Fig. 12, where we plot the ratio of apparent absorption coefficient and the scattering coefficient as a function of *ATN*. We see two features: firstly, the cross sensitivity of absorption to scattering is small in the range of below 1% to 1.5%, smaller than estimated for older Aethalometers (Rosen and Novakov, 1983). Secondly, the cross-sensitivity is almost constant over the whole experimental range and does not depend significantly on *ATN*. Cross-sensitivity does have some dependence on the size of the absorbing particles. For the particle soot absorption photometer (PSAP) Nakayama et al. found a size dependent sensitivity. We have used absorbing particles of different size to challenge the response of the Aethalometers AE33 and found that *C* does not depend greatly on the absorbing particle size within the measured range (ACTRIS, 2014b)."

We added the caption to Fig. 12:

**"Figure 12. The AE33 instrumental cross-sensitivity to scattering – the ratio of apparent absorption coefficient and the scattering coefficient as a function of attenuation (ATN). "**

The following references were added:

ACTRIS, ACTRIS Workshop on the Reference method Multi-Wavelength Absorption, 2014b, (accessed 6 March, 2015), <http://www.wmo-gaw-wcc-aerosol-physics.org/files/ACTRIS-Reference-Method-Multi-wavelength-Absorption-03-2013.pdf>

Müller, T., Laborde, M., Kassell, G., and Wiedensohler, A.: Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer, *Atmos. Meas. Tech.*, 4, 1291-1303, doi:10.5194/amt-4-1291-2011, 2011b.

Rosen, H., Hansen, A. D. A., Gundel, L., Novakov, T.: Identification of the optically absorbing component in urban aerosols, *Appl. Opt.*, 17, 3859-3861, 1978.

Rosen, H., and Novakov, T. : Optical transmission through aerosol deposits on diffusely reflective filters: a method for measuring the absorbing component of aerosol particles, *Appl. Opt.*, 22, 1265-1267, doi:10.1364/AO.22.001265, 1983.

There seems to be much confusion in the terminology relating to the conversion of optical measurements in mass concentration. We have attempted to explain the conversion going back to the original determination of the attenuation mass cross-section (Gundel, 1984). Gundel et al. (1984) are very brief on the topic of the treatment of filters to remove the non-refractory particles; we therefore drew on the fact that a coauthor of the submitted manuscript (ADAH) has been a member of the group at the Lawrence Lab conducting these measurements in the 1980's.

We have extended the discussion of this topic in the Supplement and changed the text accompanying Table S2 to:

“The mass absorption cross-section values  $\sigma_{air}$  denotes the mass absorption cross-section of aerosols freely suspended in the atmosphere. The values used in the Aethalometer assume an inverse dependence on the wavelength – that is, a “black” sample with an Ångström exponent equal to unity (Moosmüller et al., 2011).

The relationship between the absorption and mass was determined by optical measurements of transmission and thermal measurements of samples, where the non-refractory constituents of the carbonaceous sample were removed (Gundel et al., 1981; Gundel et al., 1984). The pollutants in ambient air and combustion source exhausts were sampled on quartz fiber filters and analyzed. The non-destructive optical measurement was the laser transmission method carried out at room temperature. The thermal method employed was the evolved gas analysis (EGA) in oxygen (Melissa et al., 1976). EGA was performed on the non-treated filters and on the filters sequentially extracted by benzene and a mixture of methanol-chloroform. The most refractory peak in the extracted filter thermograms was used to quantify black carbon. The mass attenuation cross section for the laser was thus obtained:  $\sigma = (23.9 \pm 2.0) \text{ m}^2\text{g}^{-1}$ . The high value is typical for the attenuation measurement setups using collimated laser beams (Gundel et al., 1984). In the first Aethalometer, using an incandescent lamp and a green band-pass filter (Hansen et al., 1984), the mass attenuation cross section was determined to be  $10 \text{ m}^2\text{g}^{-1}$  at 530 nm. In the ensuing commercial embodiment of the Aethalometer, again using an incandescent lamp without any filters, the mass attenuation cross section was determined to be  $19 \text{ m}^2\text{g}^{-1}$  (Babich et al., 2000). When the 880 nm LED source was introduced in the Aethalometers AE16, AE21, AE22, AE31 and AE42, the mass attenuation cross section at this wavelength was determined by comparison with older-type Aethalometers to be  $16.6 \text{ m}^2\text{g}^{-1}$  (Hansen, 2005).

The path length of a photon and hence the probability for the photon to be absorbed by a particle increases in the filter matrix due to the scattering of light by the filter fibers. This can be empirically taken into account using a single parameter, describing the enhancement of absorption as a multiplication factor  $C$  (Weingartner et al., 2003):  $\sigma = C \sigma_{air}$ . The same mass of the sample absorbs  $C$ -times more when the particles are embedded in the filter matrix than

when they are freely suspended in the air. The separation of  $C$  and  $\sigma_{air}$  parameters, using the value of  $C = 2.14$  (determined in Weingartner et al., 2003) for legacy type Aethalometers, such as AE31, determines the mass absorption cross-section of freshly emitted BC  $\sigma_{air} = 7.77 \text{ m}^2\text{g}^{-1}$ . We use this parameter in the AE33 in addition to the parameter  $C$ , determined for the new TFE-coated glass fiber filter tape (see section "3.3 Influence of the filter material"). We measured ambient BC concentrations using collocated Aethalometers AE31 and AE33, then determined the parameter  $C$  for the AE33 from these ambient data.

Optical measurements in the infra-red part of the spectrum should be used to convert the optical measurement into the mass concentration, as the contribution of sample components other than black carbon is negligible at these wavelengths (Sandradewi et al., 2008a; Sandradewi et al., 2008b; Fialho et al., 2005; Yang et al., 2009; and references therein). The relationship between the mass concentration of BC and the optical absorption can be determined by comparing the filter photometer measurements with those obtained by thermal-optical analysis (Sciare, 2011). However, as the determination of elemental carbon (EC) depends on the thermal-optical analysis method, sometimes with large differences (Bae et al., 2009), the determination of the mass absorption cross-section also depends on the thermal-optical method employed. Additionally, the season and the sample composition (Bae et al., 2009; Chiappini et al., 2014) may influence the determination of EC; and the mass absorption cross-section may depend on the aerosol mixing state and size (Bond and Bergstrom, 2006). Concurrent determination of EC in filter samples with an thermal-optical method, and the Aethalometer measurement of optical absorption allow the site-specific determination of the mass absorption cross-section, which is specific to the thermal-optical method employed – this procedure is often employed in source apportionment campaigns, where mass closure is attempted (Sciare, 2011)."

We have added the references in the Supplement:

Babich P., Davey M., Allen G., Koutrakis, P.: Method comparisons for particulate nitrate, elemental carbon, and PM<sub>2.5</sub> mass in seven U.S. cities, *J. Air Waste Manag. Assoc.*, 50, 1095–1105, 2000.

Gundel, L. A., Dod, R. L., Novakov, T.: Determination of black carbon by thermal analysis, in Chapter from the Energy and Environment Division, Annual Report 1980, LBNL Paper LBL-11986, 5-26–5-28, 1981.

Hansen, A. D. A., Rosen, H., and Novakov, T.: The aethalometer – an instrument for the real-time measurement of optical absorption by aerosol particles, *Sci. Total Environ.*, 36, 191–196, 1984.

Hansen, A. D. A., *The Aethalometer Manual*. Magee Scientific, Berkeley, California, USA, 2005.

Malissa, H., Puxbaum, H., Pell, E.: Zur simultanen relativkonduktometrischen Kohlenstoff- und Schwefelbestimmung in Stäuben. *Z. Anal. Chem.*, 282, 109-113, 1976.

**Figures 2, 4, and 9: use larger fonts.**

The figure font size has been increased. The size depends on the final typesetting, which we were not aware of until the manuscript was published.

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**Anonymous Referee #4**

Atmos. Meas. Tech. Discuss., 7, C4525–C4528, 2015

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**General comments:**

*English syntax is understandable but not always smooth and readable. This should be reviewed and improved throughout the manuscript. There is frequent mention of source apportionment but no actual source apportionment was done for this study and manuscript. The loading compensation results and the resulting improvement in wavelength dependence of absorption may be important for source apportionment since the accuracy of the results may be improved and uncertainty reduced. A presentation and discussion of the uncertainty in the BC, babs and absorption Ångström exponents with and without the two-spot, real-time filter loading correction is needed in the context of source apportionment. Discussion of source apportionment should be limited to this aspect of the discussion and conclusions with reference to published apportionment studies and the effect of reduced uncertainties of absorption Ångström exponent. At a minimum the effect of the two-spot loading compensation from the several sites (and events within that data) on Ångström exponent should be quantified.*

*The data, figures and tables, are not consistent with regard to using a common wavelength, defining the wavelength presented or explanation of why a particular wavelength was used in the presentation.*

*The figures and tabular information in the supplement do not materially add to the discussion and conclusions and can be summarized in a few sentences in the main manuscript. See comments below.*

We thank the referee for her/his comments and address the points she/he raised in more detail below. A native English speaker has reviewed the manuscript and we have made the necessary parts smoother and, hopefully, readable. We discuss BC, the absorption coefficient and the absorption Ångström exponent as all of these parameters have been derived from Aethalometers measurements, in context of source apportionment, climate forcing, and determination of Saharan dust events, to name a few Aethalometer application topics.

Source apportionment to which we refer to and which the referee mentions is described in Sandradewi et al., 2008b. A summary on the uncertainty of the BC concentrations and the Angstrom exponent, as determined by the new Aethalometer, is already presented in Table 2. We have expanded the discussion of this table in the section about the source apportionment to guide the reader. We have expanded and rewritten the last paragraph in section “3.2 Real-time compensation of AE33 data”, where the increase in the wood burning fraction of BC is discussed for the presented campaign and in light of other studies using this technique:

“The loading effect is stronger at lower wavelengths than at higher ones, influencing the measurement of absorption to a higher degree in blue than in the IR part of the spectrum. The loading effect therefore reduces the determined absorption Ångström exponent  $\alpha$ , calculated from  $\alpha = \ln(b_{470\text{nm}}/b_{950\text{nm}}) / \ln(950/470)$ , as shown by the difference between the intercept  $\alpha_0$  and the uncompensated average value  $\bar{\alpha}$  (Table 2b). Uncompensated data will not yield a correct value of  $\alpha$ , resulting in incorrect fractions when performing black carbon source apportionment. The non-compensated average Ångström exponent lies close to 1. This value is consistent with pure diesel exhaust measured at the site. After compensation of the data, the average Ångström exponent increases to 1.21 (using Aethalometer AE33 and TFE filter material). This change is significant, as the average BC apportioned to wood burning increases by  $1 \mu\text{g}/\text{m}^3$  and the average BC fraction apportioned to wood burning increases from none to 16% with a large diurnal variation (Fig. 9b). A similar underestimation of the contribution of wood burning would be observed in Grenoble, France, (Favez et al., 2010), and about a third lower than the actual contribution in Roveredo, Switzerland, (Sandradewi, 2008b), if non-compensated data had been used for source apportionment of BC. The use of the compensation algorithm greatly improves the calculation of the Ångström exponent. Consequently, real-time source apportionment is possible and has been implemented in the Aethalometer model AE33. ”

Black carbon concentration (BC) is reported from the measurements carried out at 880 nm throughout the manuscript, except for the MAAP data in Figure 11. We have reported this in section “2.5 Black carbon concentration (BC) calculation”, where we write: “BC is historically defined by Aethalometer measurement at 880 nm...” (p. 10191, line 8). We have changed the caption of Fig. 11 to (also taking into account the comment from **Referee #5** on Fig. 11):

**“Figure 11. Regression: BC measurements by AE33 (determined at 880 nm) and MAAP (determined at 637 nm). The data were resampled to 5 min for both instruments. The Aethalometer AE33 uses measurements at 880 nm and parameters  $C=1.57$  and  $\sigma_{\text{air}}=7.77 \text{ m}^2/\text{g}$  to obtain BC. MAAP determines BC at 637 nm using  $\sigma_{\text{air}}=6.6 \text{ m}^2/\text{g}$ , the effective parameter  $C$  is not disclosed and is inherent in the instrumental algorithm.”**

Supplement data shows the importance to perform filter loading effect compensation. There is no published data showing systematic differences in filter loading effect between different sites. This is the motivation for the new instrument: the effect is variable and should be determined for each measurement site and with high temporal resolution. We think this warrants the inclusion of the topic in the Supplement. Furthermore, due to comments of the other reviewers, we extended the Supplement and more often refer to the latter within the manuscript.

**Further comments by page and line.**

**P10183, line 15**, *The loading effect has been shown in fact, not just possibly, to depend on absorbing aerosol properties such as size distribution and ageing of the black carbon particles. D. Lack et. al.*

We have modified this sentence in accordance with the comments by Referee #1 and #4, please see the answer to the Referee #1's comment, **P10183 L15-16**. The interpretation of the compensation parameter and its relation to ageing of BC-containing particles is a topic of our current research (Drinovec et al., 2013).

**P 10185 line 10** *Is the third significant digit 7.77 valid and needed? The value 7.7 is used later. This applies to the table in the supplement as well. Reference this number absorption cross section number.*

We have been presented with the same dilemma when writing the manuscript – and we share the referee's feeling that reporting the mass absorption cross-section to four digits is sometimes an over-kill. However, in the large volume of literature, two decimal places have been used, sometimes resulting in reporting four “relevant” digits, which becomes relevant when reporting high urban BC concentrations. With the default measurement configuration, the precision of the measurement lies at 30 ng/m<sup>3</sup>. The third digit is necessary to avoid a systematic error at background sites when performing calculations using different wavelengths, for example determination of the wavelength dependent SSA at Jungfraujoch (Collaud Coen et al., 2004). We therefore use 7.77 throughout the manuscript.

**Line 20** *ATN and ATNmax should be referenced to the basic eqn. (3) in section 2.4.*

We have added the reference to Eq. (3) (now Eq. (1), please see below, **P10186, line 19**) in the text.  $ATN_{max}$  was changed to  $ATN_{TA}$  for consistency within the manuscript. Please see also the answer to **Referee #5, P. 10185; L. 20**.

**P10186, line 1 and 2, and figure 1** *The details of figure 1 need more explanation. What controls the flow through filter S1? A mass flow meter is shown but no valve or orifice is shown in that flow path. Are the flows critical at the orifices? What are the “different modes of operation” and how do the valve positions relate to them? Alternatively, since modes are not mentioned further, and figure 1 is schematic, it could be simplified. See comment to P 10190 below.*

We have modified Figure 1, adding a table describing different modes of operation, and expanded the caption of Figure 1 to maximize the information:

“Figure 1. The AE33 flow diagram. During measurement, the inlet air passes through filter spots S1 and S2, each with a different flow rate, as set by the orifice 2. Airflow through S1 is measured by the mass flowmeter 1, flow through S2 is calculated as a difference between the total flow (flowmeter 2) and flow through S1. The valves are used for routing of airflow during different modes of operation (see the table). The bypass mode is used during the tape advance procedure with orifice 1 mimicking the filter flow resistance.”

*line 19 section 2.3 and 2.4 following Without equations presented later, equation 1 and the associated discussion are not clear. The basic equation, eqn. 3, should be presented first then expanded with terms to account for the various additional terms and corrections including the filter loading effects, both past from references and the present results.*

Section 2.3 describes an empirical observation: the non-compensated  $BC$  linearly depends on  $ATN$  and that the slope can be used as a criterion whether loading effects are present in the data. This concept is discussed, including the implicit assumptions made when plotting  $BC(ATN)$ , and extended to other parameters, particularly the absorption Ångström exponent.

Section 2.4 starts with the description of the loading effect and shows that the linear behavior of the  $BC(ATN)$  plot is consistent with the parametrization of the loading effect with Eq. (7). We then proceed to using the “two-spot” approach to show how to measure the loading effect. We have extended the introduction of the manuscript to prepare and guide the reader along these lines, by expanding and rewriting the last paragraph:

“A solution, which is superior to the compensation methods described above, is to measure the nonlinearity itself with high time resolution by the filter photometer itself. First, we start by introducing the loading effect and the method to measure it from the non-compensated data. Then we describe an approach to show how to measure the loading effect. This is achieved by measuring the attenuation of light on two sample spots with different loading, and using this information to extrapolate the measurements to zero loading, thereby eliminating the non-linearity. We introduce the new Aethalometer model AE33 that performs such a measurement and compensates for the nonlinearity in real-time.”

We have moved the former Eq. (3), now Eq. (1), to the start of section “2.3 Evaluation method for loading effect compensation algorithms”. References to equations were appropriately changed.

**P 10187 line 22** *Is the supplemental relative slope,  $BC(ATN)$  analysis in Table S2 better or more comprehensive than that of Virkkula? If so, it should be summarized somewhere in the main article in terms of mean value and range over the five sites. The table is not needed.*

Table S1 (!) shows the average filter loading effect as determined with the  $BC(ATN)$  analysis for different measurement sites and seasons. The point we wanted to make is that the effect is variable and should be determined for each measurement site and season. The relationship



between the *BC(ATN)* slope and the Virkkula compensation parameter is explained in the response to **Referee #1's** comment, **P10188, L24-25**. The parameters are the same to the first order. In the AE33 operated with quartz and TFE-coated fiber tape the compensation parameters differ slightly, as shown in Figures 6 and 7 (Sections 3.2 and 3.3) because of the different tape material. Looking at the Virkkula parameter determined for the Aethalometers AE31, one should consider these differences when comparing it to the compensation parameter determined automatically (and with a higher time resolution) in the AE33.

**P 10188 line 9** *What is meant by “started to saturate”?*

Start of the saturation is meant to describe the deviation from the linear behavior. This has been clarified, please see the answer to **Referee #1's** comment, **P10188, L8**.

**P 10190** *The meaning of lateral air flow in the chamber is not clear from the discussion or the flow diagram fig. 1. What is lateral air flow in this case? Is it the lateral air flow within the filter matrix? Is mass flow converted to volumetric at atmospheric temperature and pressure or better yet STP? Specify the flow standard in discussions.*

This is indeed flow within the filter matrix, we have added this description to the text, please see the answer to **Referee #1's** comment, **P10190 L14-15**. The flow standard in the Aethalometers can be volumetric, STP or a user setting. We strongly advocate reporting mass flow at standard conditions. We added the information on flow to the last paragraph in section 2.1:

*“The airflow is measured by two mass-flowmeters after the aerosol particles are deposited on the filter. The flow can be reported at ambient conditions, conditions set by the users, or different standard conditions, the default being 20.11° C and 1013.25 hPa.”*

**P 10191** *The table S2 and caption in the supplement can be eliminated and summarized here as “based on  $7.8 \text{ m}^2 \text{ g}^{-1}$  and Ångström exponent of one.”*

To maximize the information conveyed in the manuscript and to keep all information about the new Aethalometer in one publicly accessible, refereed publication, which can be referenced, we have decided to keep Table S2. Please see also reply to **Referee #1, P10191 L8-12**.

**P10192** *Provide more details about the TROPOS site. The general weather condition statements can be deleted unless they are germane to and used in the analysis of the data and loading compensation.*

We have observed that the compensation parameter  $k$  may be interpreted in terms of aerosol age (Drinovec et al., 2013). The interpretation of the compensation parameter and its relation to ageing of BC-containing particles is a topic of our current research (Drinovec et al., 2013). High winds may have brought more regionally representative (and hence aged) aerosols to the measurement site; therefore we mention this in the manuscript. We have expanded the last paragraph in section 2.6 and added the reference to the description of the site:

“During the campaign, the weather was cloudy and windy with temperatures slightly below zero (ACTRIS, 2014a). The measurement site is generally considered to be moderately polluted urban background influenced by a mixture of distributed sources and some closer roads (Wehrner and Wiedensohler, 2003), but the winds may have mixed the atmosphere so that the site could be representative of a broader region.”

We have added the reference:

Wehner, B. and Wiedensohler, A.: Long term measurements of submicrometer urban aerosols: statistical analysis for correlations with meteorological conditions and trace gases, *Atmos. Chem. Phys.*, 3, 867-879, doi:10.5194/acp-3-867-2003, 2003.

**P10192 line 25** *Section 2.7 is redundant with previous mention of source apportionment.*

It is true, we could have referenced the source apportionment model as: Sandradewi et al., 2008b. To provide more information on the source apportionment model for the non-specialists, we chose to include a short summary. In addition, we report in section 2.7 the parameters used for the source apportionment model.

**P110199** *This is not a conclusion unless based on the experimental or model results. The improvement in spectral dependence would need to be applied to past source apportionment models to quantify and document the improvement in source apportionment. “Improvements in the determination of the spectral dependence of absorption, as described by the Ångström exponent, allows for real-time high time resolution source apportionment by the AE33.”*

This relates to the comment also mentioned in the Referee’s **General comments**. We report the difference in the determined absorption Ångström exponent in Table 2b. We have expanded the section “3.2 Real-time compensation of AE33 data” to further explain the consequences of using non-compensated data, and the low bias to the determination of the wood burning fraction of BC due to loading effects. Please see our response to the reviewer’s **General comments** above.

**Table 4** *What were the two or three AE33 instruments used for the first set of correlations? Was this part of the Klagenfurt and TROPOS experiments or an external experiment, i.e. USEPA?*

The campaign (ACTRIS, 2014a) included several older type Aethalometers (AE31, AE22) and several new ones (AE33). The comparison referenced as ACTRIS (2014a) was indeed conducted at TROPOS. We have added the information in the table caption to elucidate the locations of the tests. An additional reference was added to the Table 4:

**“Table 4. The slope and  $r^2$  of the linear least square regression through zero of BC measurements from different filter photometers. The campaigns were conducted in: Leipzig at TROPOS (ACTRIS, 2014a); Columbus, Ohio, USA (USEPA, 2014); the Aerosol d.o.o. aerosol chamber; Borgerhout, Belgium (Maetz et al., 2013); ZF2 ecological reserve, 55 km north of Manaus, Brazil (Holanda et al., 2014).**

|      | slope relative to AE33 | $r^2$ | comparison time resolution | test duration | reference              |
|------|------------------------|-------|----------------------------|---------------|------------------------|
| AE33 | 1,03                   | 0,99  | 5 min                      | 2 days        | ACTRIS, 2014a          |
|      | 0,96                   | 1,00  | 5 min                      |               | ACTRIS, 2014a          |
|      | 1,06                   | 0,99  | 1 hour                     | 30 days       | USEPA, 2014            |
| AE31 | 1,05                   | 0,95  | 5 min                      | 1 day         | ACTRIS, 2014a          |
|      | 0,84                   | 0,94  |                            |               | ACTRIS, 2014a          |
|      | 1,02                   | 0,93  |                            |               | ACTRIS, 2014a          |
|      | 0,93                   | 0,92  |                            |               | ACTRIS, 2014a          |
|      | 1,10                   | 0,91  |                            |               | ACTRIS, 2014a          |
|      | 1,13                   | 0,90  |                            |               | ACTRIS, 2014a          |
| AE22 | 0,93                   | 0,92  | 5 min                      | 1 day         | ACTRIS, 2014a          |
|      | 1,07                   | 0,99  | 5 min                      | 2 weeks       | chamber, this study    |
|      | 0,99                   | 0,93  | 1 day                      | 3,5 months    | Maetz et al., 2013*    |
| MAAP | 0,96                   | 0,98  | 5 min                      | 3 days        | ACTRIS, 2014a          |
|      | 1,07                   | 0,97  | 30 min                     | 4 months      | Holanda et al., 2014** |

\* Maetz et al., 2013, compensated the AE22 data using Virkkula et al., 2007.

\* Holanda et al., 2014, PM2.5 inlet.”

**Figure 4** What is the abscissa, not labeled, in figure 4b? Is this binned data from figure 4a? Figure 4a has a significant amount of babs data less than 100 inverse megameters while 4b shows very little in that range.

The figures have been modified with the addition of labels and font sizes increased (please, see also our reply to **Anonymous Referee #1, Figures 2, 4, and 9**). Data in Fig. 4b shows binned data,  $b_{abs}$  averages in bins 1 ATN unit wide, as explained in the figure caption – this is also the reason for the smaller number of points in Fig. 4b below  $100 \text{ Mm}^{-1}$ . With lower concentrations, the ATN changes more slowly, resulting in more measurements (time-series shown in Fig. 4a) being included in one bin. An example of the concentration distribution in one bin is shown in Fig. 2 (with a different bin width).

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### Anonymous Referee #5

Atmos. Meas. Tech. Discuss., 7, C4596–C4599, 2015

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*This paper presents the operating principle of a new version of a commercially available Aethalometer where the measurement artefact caused by the filter loading effect is continuously measured. The instruments firmware corrects the measured values for this systematic error.*

*My main criticism is that this paper focuses predominantly on the filter loading effect. I agree that the new instrument can correct for this bias, but the fundamental weaknesses of the Aethalometer are neither addressed nor is it carefully discussed. The paper is not balanced, and it appears that it was written from the “manufacturer perspective” with the goal to promote the new feature of this instrument.*

We thank the referee for her/his comments and address the points she/he raised in more detail below

The aim of this manuscript was to fully disclose the new compensation algorithm, as implemented in the new Aethalometer, to the academic public. To make the approach totally transparent a conflict of interest for certain authors was disclosed in the acknowledgement section. We further call on to the Editorial board of ATM(D) to mandate the disclosure during the submission process.

*It should be made clear that the correction of the filter loading effect does not solve the basic problem of this method. The main uncertainty in the derived values (measured light absorption coefficient or black carbon concentration) is caused by unknown multiple scattering of light in the filter. The optical path within the filter is larger and therefore the particle’s absorption is principally overestimated. This correction (expressed as a factor C) is much greater and more uncertain than the presented correction of the loading effect. According to the literature the correction factor for this enhanced absorption lies between 2 and 6. The C-values are dependent*

*on various factors such as the filter material, the amount of light scattering particles embedded in the filter, the mixing state of scattering and absorbing particles, the face velocity, relative humidity, etc. The wavelength dependence of  $C$  is unknown, and it is often assumed to be constant. Therefore, more discussion on the uncertainty related to  $C$  (and MAC) should be given in the paper.*

*It is necessary to present all assumptions in the calculation of BC. You may argue that eq. 6 gives the answer, but there the discussion is passed on the definition of parameter sigma which is not well introduced and contains two unknowns ( $C$  and MAC). Please address all these uncertainties and mention all assumptions (e.g. ignoring the wavelength dependence).*

*For curiosity: unpublished studies found indications that the two artefacts depend on the instruments flowrate. An explanation is that the depth, at which the particles are incorporated into the filter matrix, depends on the filter face velocity. Did you find such indications with the new instrument? If yes, this would mean that it is important to assure that the face velocities are equal on both filter spots (which is not the case).*

The article deals with the filter loading effect and the dual spot compensation algorithm as the article title suggests. The parameter  $C$  is relative to the method one uses for the comparison (please, see the response to **Referee #1, P10191 L1-2 and L8-12**). The prerequisite for the determination of the multiple scattering parameter  $C$  it is crucial to first compensate the data for the filter loading effect, otherwise the determination of  $C$  suffers from the loading effects. Determination of  $C$  for the TFE-coated glass fiber filter used in dual-spot Aethalometer is part of on-going work (ACTRIS, 2014b) and will be a focus of future article (in preparation). The influence of the filter tape material is discussed in section “3.3 Influence of the filter material”. We investigated the influence of the face velocity on parameter  $C$  and we also investigated measured BC concentrations for both spots at small filter loadings; difference in  $C$  between the spots (having different face velocities) at low  $ATN$  would result in  $FVRF$  being different from 1, which was not the case. For the details on the influence of size and face velocity and scattering material on the filter on the instrumental response, please, see the response to **Referee #1, P10189, L15, P10191 L1-2 and L8-12**. The influence of the mixing state on the compensation parameter  $k$  is a topic of on-going work (Drinovec et al., 2013, and Drinovec et al., 2014b).

#### **Other issues:**

*The Aethalometer measures an apparent absorption coefficient which is translated into an apparent black carbon and – in a next step – into a “true” black carbon mass concentration. In the paper, only the term BC is used and I emphasise that it is clearly stated where BC was corrected and were not (as in Virkkula et al., 2007). I further recommend to adopt the nomenclature presented in Petzold et al., ACP, 2013.*

We have modified Figure 2a and 2c and now use  $BC_{NC}$  or BC-nc. for non-compensated measurements of BC; and BC and BC-comp. for compensated ones in all figures and tables.

Please see the answer to **Referee #5 P10186, line 19** for more details. We do use the most recent nomenclature (Petzold et al., 2013), in the introduction, we write (page 10182, line 13):

“The attenuation coefficient is converted to the absorption coefficient, and the mass equivalent black carbon concentration (Petzold et al., 2013; we will use BC below as a short form) is calculated by dividing the absorption coefficient with the BC specific mass absorption cross-section.”

**Page 10185; Line 10:** *The MAC Value of 7.77 m<sup>2</sup>/g is low. The former instrument used an apparent MAC value of 16.6 m<sup>2</sup>/g at 880 nm. Where does this difference come from? Is 7.77 m<sup>2</sup>/g not the apparent value? Again: a description is needed which presents all assumptions and shows how this MAC value is incorporated into the calculation of BC. How was b<sub>abs</sub> (Fig 4) calculated (C-value)?*

The new Aethalometer AE33 uses the same  $\sigma = C \sigma_{air} = 16.6 \text{ m}^2/\text{g}$  value as the older type Aethalometers. Because of the different filter material, the value of parameter C is different, as discussed in “3.3 Influence of the filter material”. Please, see the detailed response to **Referee #1, P10191 L8-12**.

**P. 10185; L. 20:** *“When the attenuation reaches a certain threshold...” at which wavelength?*

Attenuation (ATN) increases with the falling wavelength, so larger ATN values are measured at lower wavelengths. The threshold is therefore always reached first at 370 nm. We have modified the text to:

“When the attenuation reaches a certain threshold, a tape advance is induced so that measurements start on a clean spot. The attenuation threshold, first reached at the lowest wavelength 370 nm, is called  $ATN_{TA}$  (see Eq. (1) in section 2.3 below) with a default value of 120, which can be changed by the user – for example to conserve the filter tape by setting the value larger.”

**P. 10186; L. 6:** *Mention here that changing the filter type is expected to influence the optical filter properties and therefore change the corrections (both, the multiple scattering and loading artefact).*

We have added the sentence:

“Different filter types influence the loading effect and the multiple scattering in the filter matrix differently.”

**P. 10187; L. 4:** *Looking at Fig. 3: it’s not true that BC(ATN) can well be approximated with a linear fit. Why?*

We believe the referee correctly wanted to point out that the reference on p. 10187, line 4 to Fig. 2b is wrong. A reference to Fig. 2a should be used here instead of Fig. 2b. Figure 2a shows that inside the fitting range a linear fit is an excellent approximation of the measured reduction of sensitivity. We changed the reference accordingly. Figure 3 shows the comparison between different compensation models.

**P. 10195; L. 24:** *I do not clearly understand how C was determined. For this you need to compare the Aethalometer measurement to an independent reference (ideally measuring true absorption). As this was not available in Klagenfurt, I have to assume that you rely on to  $C=2.14$  for quartz filters. As this value has uncertainties ( $C$  ranges from 2 to 6), only ratios of  $C$  for different filter materials can be reported. Please clarify.*

Parameter  $C$  for the TFE coated glass filter was set to give the same values of  $BC_0$  (the intersect in the  $BC(ATN)$  plot (Fig. 2a and Table 2), please see section “3.3 Influence of the filter material”) for both filter types. For the TFE coated glass filter, we obtained:  $C \approx 1.57$  using the 2 week Klagenfurt campaign data. The  $C$  value of 1.57 was indeed calculated assuming that value 2.14 for the quartz filter. For a long discussion concerning the parameter  $C$ , please see the response to **Referee #1, P10191 L8-12**. Additionally, the last sentence of the first paragraph in section 3.3 was extended with:

“For the TFE-coated glass filter we obtained:  $C \approx 1.57$  using the 2 week Klagenfurt campaign data, assuming  $C=2.14$  for quartz filter (Weingartner et al., 2003). There is some uncertainty with this value since values of  $C$  for quartz filter as obtained by comparisons with other techniques differ (Weingartner et al., 2003; Collaud Coen et al., 2010; Müller et al., 2011, Segura et al., 2014).”

**Fig. 1:** *How is the flow through S1 controlled? It is unclear to me what orifice 1 is good for.*

The flow ratio  $F2/F1$  is controlled by the orifice 2. Orifice 1 is used to increase flow resistance during the filter bypass procedure. An updated flow diagram is now shown and different operation modes are explained in detail in Fig. 1. Please check our answer to **Referee #1 P10186, line 1 and 2, and figure 1**.

**Fig 2:** *in a) BC conc. in the ATN range 25-30 is approximately 4000 ng/m<sup>3</sup>. The corresponding peak in c) is at 2000 ng/m<sup>3</sup>. This is not consistent.*

The distribution of BC is asymmetric with an average value of 3886 ng/m<sup>3</sup> and a median of 2831 ng/m<sup>3</sup>. The  $BC(ATN)$  plot presents BC averages in each bin. The apparent discrepancy results from different statistical approaches.

**Fig 2&3:** I understand that BC was calculated using 880 nm. Therefore, it makes sense that the ATN values in Fig 2 range up to 60. Why are ATN values in Fig 3 range up to 120? Is this for a different wavelength? Please clarify.

Figure 3 illustrates a general filter loading effect for any channel. Since the maximum attenuation is measured in the UV (370 nm), we use this limit to plot the model curves. Please see also the reply to **Referee #5 P. 10185; L. 20** just above.

**Fig 3:** The green curve has a slight curvature. But: the Virkulla correction is linear and therefore the green curve should be a line.

Virkkula (2007) uses the following compensation equation:  $BC = BC_{NC} \cdot (1+k \cdot ATN)$ . The relative sensitivity thus follows a hyperbolic curve:  $BC_{NC}/BC=1/(1+k \cdot ATN)$ .

**Fig 11:** The interpretation of the slope makes no sense as long as these values are not known: sigma or MAC value used by the MAAP, MAC value of the Aethalometer, C-Value. Also indicate the wavelength of the measurements as these values are wavelength dependent.

Measurement wavelengths,  $\sigma_{air}$  and C values are included in the figure text except for the MAAP, where the algorithm to calculate absorption algorithm is not fully disclosed along with the value of C (Petzold and Schönlinner, 2004; Petzold et al., 2005). We have changed the caption to Fig. 11 accordingly; please see **our answer to Referee #4, General comments**.

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