Responses to Review of Referee 3 (amt-2015-33)

We would like to acknowledge to the anonymous referee 3 for his/her useful remarks and comments which have helped to improve the manuscript. All comments have been addressed as detailed in the following responses (in red).

The authors present an interesting measurement setup using two Aethalometers in parallel, each with a different flow (face velocity), causing a different deposition rate and consequently different attenuation of light through the filter tape loaded with particles. The concept has been used in the newly developed Aethalometer AE33 (Drinovec et al., 2014). The approach presented in the reviewed manuscript uses a simple linear model to determine the factor k, which accounts for the loading effects in the filter tape.

The authors assert that the presented method results in accurate determination of BC and that the results can be made independent of the aerosol scattering. They propose their method as the empirical method for the determination of BC in situations similar to the one they describe. The authors need to address several issues:

1. Accuracy

The claim that the determination of BC is accurate is not correct. The authors show that the BC's determined from the Aethalometer using 2 l/min and the one using 6 l/min are the same (Fig. 7). This is not true, what they show is that the two Aethalometers give precise results. The criterion for accuracy is missing. Given the fact that the cite Park et al. (2010) and Drinovec et al. (2014), they could have used the algorithms described there to determine the accuracy. This should be added to the manuscript. It would be highly advantageous for their argument if they showed the slopes in the regression plots and mention them in the text. The uncertainty (or confidence interval) of the fit parameters should be shown as well.

Response:

The fitting results have been added in Fig. 7 (now it is Fig. 8 in the revised manuscript). The objective of this study is to minimize the loading effect on the BC measurement results.

2. Definition of attenuation

Attenuation (ATN), as defined in Eq. 1, is a measure of how much light is transmitted though the filter. The subsequent definition of the "corrected" or "true" ATN is, simply put, wrong! There is no "wrong" or "right" ATN, and no "corrected" one either – ATN is defined in Eq. 1. The under-reporting of BC due to loading effect in

the filter is not due to the measurement of ATN, but due to the non-linearities (higher order effects), which are not described by Eq. 4. The loading of the tape definitely influences the ATN measurement, it causes the ATN to increase with more and more (absorbing) particles being loaded on the filter. The relation is on-linear and additional terms need to be taken into account to convert the measurement (!) of ATN into absorption or mass concentration. Eq. 5 is therefore wrong, and needs to be rewritten so that it shows the relationship between the "raw" BC measurement and the "corrected" BC – taking a form of Eq. 3, only with BC from Eq. 4 (see also comment P2856, L7). The authors need to take this in to account in the revision of the manuscript.

Response:

1. The unsuitable terminology has been changed.
2. According to the definition of ATN, the ATN value is dependent on both I and I_0. Further, the intensity of I is affected by the amount of BC aerosol deposition.
3. The eq. (4) to (5) (eq. (4) to (6) in the revised manuscript) have been modified as:

"On the basis of the same principle as eq. (2), the average BC (or $\overline{\mathrm{BC}}$*) from initial time* t_0 *(*$t_0 = 0$*) to time t can be expressed as*

$$\overline{\mathrm{BC}} = \frac{\Delta \mathrm{ATN}}{\Delta t} \cdot \frac{A}{Q} \cdot \frac{1}{\sigma_{\mathrm{ATN}}} = \frac{\mathrm{ATN} - \mathrm{ATN}_0}{t - t_0} \cdot \frac{A}{Q} \cdot \frac{1}{\sigma_{\mathrm{ATN}}} = \frac{\mathrm{ATN}}{t} \cdot \frac{A}{Q} \cdot \frac{1}{\sigma_{\mathrm{ATN}}} \qquad (4)$$

where ATN_0 *(*$\mathrm{ATN}_0 = 0$*) is measured light ATN value at* t_0*.*

Then, eq. (3) can be written as

$$\overline{\mathrm{BC}_c} = \frac{\overline{\mathrm{BC}}}{(1 - k_a \cdot \mathrm{ATN})} = \frac{\mathrm{ATN}}{(1 - k_a \cdot \mathrm{ATN})} \cdot \frac{1}{t} \cdot \frac{A}{Q} \cdot \frac{1}{\sigma_{\mathrm{ATN}}} = \frac{\mathrm{ATN}_m}{t} \cdot \frac{A}{Q} \cdot \frac{1}{\sigma_{\mathrm{ATN}}} \qquad (5)$$

and

$$\mathrm{ATN}_m = \frac{\mathrm{ATN}}{(1 - k_a \cdot \mathrm{ATN})} \qquad (6)$$

where k_a *is a correction factor and it is determined from accumulated BC aerosols,* $\overline{\mathrm{BC}_c}$ *is corrected average BC value from initial time* t_0 *to time t. Additionally,* ATN_m *and ATN represent modified and measured ATN values, respectively."*

3. Immunity to scattering

The immunity to scattering has not been demonstrated in the manuscript – it is an interesting working hypothesis which needs to be tested. The authors need to test their method in environments with different concentration ranges and different values of single-scattering albedo. The scattering coefficient needs to be measured alongside the Aethalometer measurements. The sensitivity of the determination of the parameter k needs to be carried out. At low ATN on a unloaded piece of filter tape, Eq. 9 will feature a difference of two small numbers being divided by a difference of two small numbers. Noise in the measurement will significantly contribute to the value of the

parameter k. In urban environments this might not be as critical (although this needs to be proven!) as in clean ones, but the stability of the determination of the parameter k needs to be shown.

Response:

1. The statement about the light scattering effect on measurement result has been modified and is no more emphasized.
2. The following statement has been added in the revised manuscript.
   "*Additionally, it has to be noted that the problem of enhanced/reduced light ATN caused by light scattering was not considered in this proposed correction algorithm because this artifact is a much less important one for existing Aethalometer BC data.*"
3. The statement about the light scattering effect has been modified as:
   "*In this study, it was found that with increasing aerosol load on the filter, the influence of the light scattering behavior of the filter matrix could be mitigated. However, the light scattering caused by deposited aerosol might be still existed due to the properties of sampling aerosols at different environments.*"

The manuscript is often unclear and very sparse with information. For example, the following information needs to be added:

- Time resolution of the determination of the parameter k.

Response:

The time resolution is 5-min, and it has been described in the revised manuscript.

- Units need to be shown in all graphs.

Response:

The units have been added in all graphs.

- The assertion of wood burning contribution to BC needs some substantiation.

Response:

The estimated absorption Ångström exponent values > 2.0 also can be found in the measurement results.

- Consistent notation should be used in the figures and the text of the manuscript.

Response:

That has been checked.

- The underlying concept of Eq. 4, that is that all BC, accumulated on the filter (that is integrated from t=0), contributes to the loading, and that the parameter k is

determined from this integrated BC, is fundamental to the manuscript. This needs to be expanded and emphasized. This is the central point of the manuscript (Drinovec et al. (2014) make the same point, only with higher time resolution)!

Response:

The following statement has been added in the revised manuscript.

"*where k_a is a correction factor and it is determined from accumulated BC aerosols, $\overline{BC}_c$ is corrected average BC value from initial time t_0 to time t.*"

Other comments, by page/line numbers:

Page 2852, Line 2: "Recently" – for example, the Weingartner et al. (2003) paper was published 12 years ago, this is not recent.

Response:

"Recently" has been changed to "In fact".

P2853, L23: "…the decrease in light transmission through the aerosols on the filter is measured." – No, the measured quantity is the intensity of light transmitted through the filter loaded with particles.

Response:

The statement has been modified as:

"*Subsequently, the intensity of light transmitted through the filter loaded with particles is measured.*"

P2854, L7: "…the measured BC concentration decreases with an increase in the filter load…" – Virkkula et al (2007) have shown an increase (negative parameter k). A reference is missing for both claims in this sentence.

Response:

In Virkkula et al (2007), it is assumed that the BC concentration remains stable during the filter spot change and that the BC value measured with a lightly loaded filter is the closest to the real concentration value. However, the assumption that the BC concentration is stable in the ambient environment during the filter spot change is not always true. When the BC concentration at next spot is lower than that at previous spot, the k value will be negative. This negative k is not due to an overestimate. The BC at a lightly loaded filter is the closest to the real concentration value is also not true. The BC at a lightly loaded filter could be overestimated due to the light scattering effect.

P2854, L9: "Recently" – see above.

Response:

"Recently" has been changed to "In fact".

P2856-2858, Section 2.1: The discussion and definition of "true ATN" needs major revision, see comments above and below.

Response:

The unsuitable terminology has been changed as referee's suggestion. All "true ATN" in the manuscript have been changed as "modified ATN".

P2856, L7: "optical mass cross section" – this is mass attenuation cross-section. In fact, this is the parameter, showing "loading effect", as ATN (defined with Eq. 1) change is converted into mass.

Response:

"optical mass cross section" has been changed to "mass attenuation cross-section".

P2856, L19: "Therefore, determining the true ATN value was the main objective of this study." – No! "True ATN" does not exist, loading effects are a consequence of non-linearities between the mass loaded on the filter and the ATN – please see above.

Response:

The statement has been changed as:

"*In practice, if the modified ATN value can be found, then the loading effect on the BC measurement can be corrected from the change rate of the modified ATN ($dATN_m/dt$). Therefore, determining the modified ATN value was the main objective of this study. According to the definition of ATN, the ATN value is dependent on both I and I_0. Further, the intensity of I is affected by the amount of BC aerosol deposition.*"

"*In the absence of the loading artifact, the ratio of ATN values obtained at two different flow rates should be equal to the ratio of the two different sampling flow rates, and then a same BC concentration can be acquired from different sampling flow rates. Actually, the ratio of ATN values measured by the two Aethalometers was not the same as the ratio of the sampling flow rates of the Aethalometers because the aerosol loading effects varied with the aerosol deposition rate. That is, the BC measurement results were not same between these two Aethalometers. If a modified ATN value can be appropriately obtained according to different aerosol deposition rates, then BC measurements can be corrected by using the change rate of the modified ATN.*"

P2858, L24 – P2859, L1, discussion about drying and data processing: Using a dryer to reduce RH is a good and recommended practice. Since the RH did not change in winter compared to summer, not using the dryer in winter could have led to large

variations of RH in the sampling lines due to operation of the air conditioner in the shelter. This might need more explanation. More importantly, negative values in the data could be noise, induced by cycling of the RH (as induced by the air conditioner). The noise in this case is a combination of a negative spike, upon the occurrence of the perturbation, and a positive one when the conditions return to the initial ones. A positive/negative pair is also possible. Removing negative values is therefore wrong – the data need to be averaged with a window, long enough to take into account the air conditioner cycling!

Response:

1. The following statement has been added in the revised manuscript.
   "*According to the meteorological data, the dew point temperature of the sampling air was lower than the indoor temperature of the sampling cabin and it indicated that water vapor condensed on the deposited aerosols would not be happened in the winter season. Therefore, the diffusion dryer was not installed on the sampling line during the winter sampling period.*"
2. The following statement has been added in the revised manuscript.
   "*These negative BC values were caused by the analytical sensitivity of the instrument at very low ambient concentrations. If negative BC values appeared in the data set of a sampling spot, the entire data set for the sampling spot was excluded from further treatment to avoid the experimental bias occurred.*"

P2859, L12-20: Fitting method needs to be specified.

Response:

The following statement has been added in the revised manuscript.
"*A linear regression method was applied to analyze the relationships between BC concentrations measured by the two Aethalometers at two sampling flow rates.*"

P2859, L24: Why does ATN in Fig. 2 extend to very large values? ATN=400 in the channel used to calculated BC (880 nm) represents a completely black filter. ATN at which the tape advance is triggered automatically is usually at ATN=120 in the detection channel at 370 nm.

Response:

In this study, two Aethalometers were used to measure BC at different flows. The filter tape in these two Aethalometers was shifted automatically to expose a pristine spot on the filter at the same time every eight hours to ensure that the two sampling spots had a fixed starting and ending time of the sampling for internal comparison. When ambient BC was high, the measured ATN could rise to over 300 after 8 hr. And, the modified ATN could be high as >400.

P2860, L6: "…value could fall to 3 : 1." – This is unclear and needs to be explained in more detail.

Response:

The statement has been modified as:

"*The ratio of the modified ATN_{F6} value to the modified ATN_{F2} value could be close to 3.*"

P2860, 12-17: The Drinovec et al. (2014) compensation parameter k is not calculated according to the Eq. 10 presented here! This comparison needs to be completely revised. However, the comparison show identical results. Comparison with the Weingartner et al. (2003) algorithm shows identical results as well. Therefore the necessity of the second Aethalometer is questionable, if a post-processing scheme, such as Weingartner et al. (2003), is shown in this manuscript to be applicable and produces identical results.

Response:

1. The following statement has been added in the revised manuscript.
   "The model of Drinovec et al. (2015) is presented in eq. (3), and the correction factor, *k*, can be easily estimated from the following equation on the basis of the measurement results of BC and ATN at sampling flow rates of 6 and 2 L min^{-1} without a complex tuning procedure which suggested by Drinovec et al. (2015)."
2. The parameter *f* in the correction model of Weingartner et al. (2003) is significantly dependent on the aerosol type, and it was difficult to determine from field sampling data. In this study, the corrected BC was computed with different *f* values to find out the best comparable results.

P2861, Section 3.2: This section needs to significantly revised using scattering measurements or changed completely. Most of the text is speculation nor, at best, a working hypothesis – see above for more.

Response:

1. The statement about the light scattering effect on measurement result has been modified as:
   "*In this study, it was found that with increasing aerosol load on the filter, the influence of the light scattering behavior of the filter matrix could be mitigated. However, the light scattering caused by deposited aerosol might be still existed due to the properties of sampling aerosols at different environments.*"
2. The following statement has been dropped in the revised manuscript.
   "*These observation results indicated that the proposed model could overcome the*

problem of enhanced light ATN resulting from light scattering at a new sampling spot, without using any light scattering coefficient."

P2862, L24: "…to 1.17+/-0.44 (estimated from corrected ATN)…" – From which Aethalometer 2 l/min or 6 l/min?

Response:

The following statement has been added in the revised manuscript.

"*After ATN modified, the absorption Ångström exponent value evaluated from modified* ATN_{F6} *was nearly same as that evaluated from modified* ATN_{F2}*.*"

P2863, L3: Same as just above.

Response:

The following statement has been added in the revised manuscript.

"*After ATN modified, the absorption Ångström exponent value evaluated from modified* ATN_{F6} *was nearly same as that evaluated from modified* ATN_{F2}*.*"

P2863, L14: The discussion of Fig. 5 needs to include more quantitative information, for example the value of k at large ATN/Q. Is Fig. 5 essentially Fig. 4 with a log scale? Why is the ATN/Q not extended to larger values?

Response:

1. Yes, the Fig. 5 is essentially Fig. 4 with a log scale to express more detail k_a at large ATN/*Q*.
2. The ATN/*Q* at ~60 is enough. At ATN/*Q* = 60, ATN is large as 300 when Q = 5 L min^{-1}. Usually, the tape advance is triggered automatically at ATN=100~120 in the detection channel at 370 nm to avoid critical loading effect. Moreover, the Figs. 4-6 are analyzed from fitting equations based on all 5-min measurement data. The relationship between measured ATN_{F6} and ATN_{F2} can be fitted with a power law equation based on all 5-min raw data, and the fitting results are shown in Table 2 (new table in the revised manuscript).

P2863, L14: prediction of the parameter k. this is interesting, but an empirical relationship might hold for this particular site and Aethalometer only. The authors suggest that these relationships can be applied more widely – this needs to be shown, by using them somewhere else.

Response:

The following statement has been described in the manuscript.

"*Although this simple correction scheme is dependent on the aerosol type, it can be used to correct BC data when the primary source of BC and the weather conditions*

are similar to those in this study."

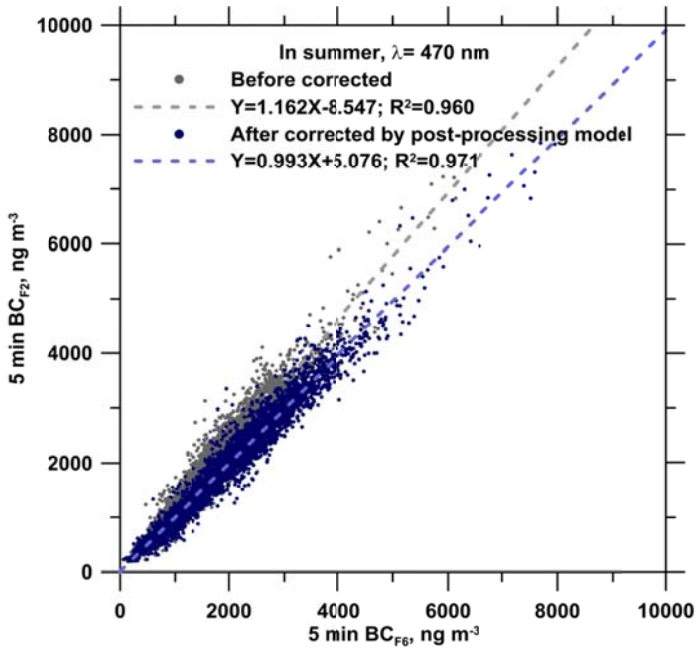
P2864, L6: The discussion of Fig. 7 needs to include the slopes. Figure showing the relationship for the measurement channel 470 nm needs to be added – it was used to determine the Angstrom exponent!

Response:

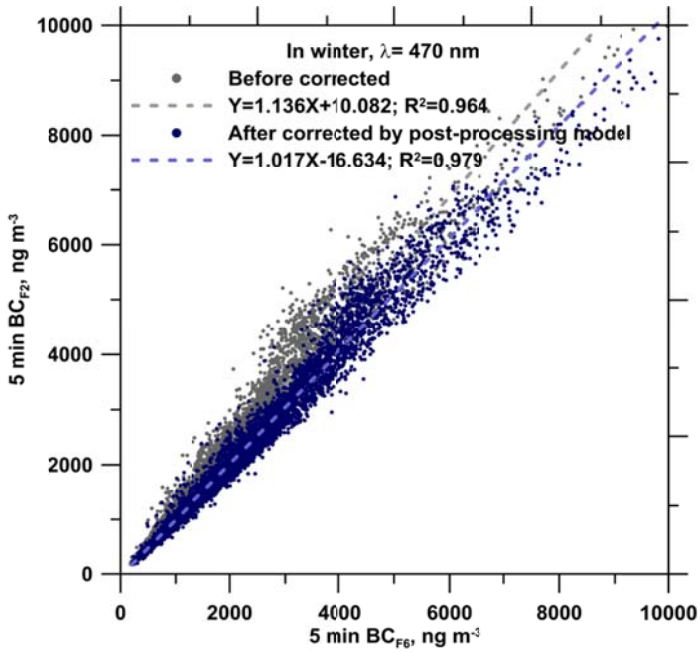
The fitting results have been added in Fig. 7 (Fig. 8 in the revised manuscript).

The figures for other wavelengths have been shown in the supplement material. They had similar results with the Fig. 7 (Fig. 8 in the revised manuscript) for other channels. Following fig. is for wavelength= 470 nm.

(a)



(b)



P2864, L11: the fact that the two measurements agree is not necessarily the proof that the derived BC value is accurate – the criteria for accuracy are missing and should be included here. Please see above for more.

Response:

The fitting results have been shown in the Fig. 7 (Fig. 8 in the revised manuscript).

P2864, L22: How were the parameters f for the Weingartner et al. (2003) algorithm determined? This is crucial for the discussion. Also, the results are identical, although the slope in Fig. 8 is missing. Please include more quantitative information (slopes…) in the discussion and in Fig. 8.

Response:

The parameter *f* in the correction model of Weingartner et al. (2003) is significantly dependent on the aerosol type, and it was difficult to determine from field sampling data. In this study, the corrected BC was computed with different *f* values to find out the best comparable results. Therefore, the slopes presented in Fig. 8 are close to 1.

The fitting results have been shown in the Fig. 8 (Fig. 9 in the revised manuscript).

P2865, L1: Same as just above.

Response:

The parameter *f* in the correction model of Weingartner et al. (2003) is significantly dependent on the aerosol type, and it was difficult to determine from field sampling data. In this study, the corrected BC was computed with different *f* values to find out the best comparable results.

P2865, L2 and onwards: Please add the discussion on the potential differences of post-processing the data with Weingartner et al. (2003) algorithm and the principle presented in the manuscript using two Aethalometers. Advantages and disadvantages should be presented, including the obvious ones (need for more instruments).

Response:

The major disadvantage of post-processing with Weingartner et al. (2003) algorithm is how to determine the parameter *f*. The parameter *f* is significantly dependent on the aerosol type, and it was difficult to determine from field sampling data. Moroeover, the post-processing model proposed in this study is also dependent on the aerosol type. However, it could be determined using two existed Aethalometers at different environments.

The following statement has been described in the manuscript.

"*Moreover, the parameter f in the correction model of Weingartner et al. (2003) is significantly dependent on the aerosol type, and it was difficult to determine from field sampling data.*"

"*Although this simple correction scheme is dependent on the aerosol type, it can be used to correct BC data when the primary source of BC and the weather conditions are similar to those in this study. Moreover, two existed Aethalometers under appropriate flow control can be used to create correction schemes for different environments.*"

P2865, Conclusion: Conclusion needs to be revised according to the comments above. The message about scattering and wide applicability needs to be either toned down or experimentally proven. Please see above for more information.

Response:

The conclusion section has been modified.

"*The proposed correction model can overcome the aerosol loading effect on BC measurement results, and it can be used in a newly designed instrument to determine the BC concentration for a minimizing artifact in real time by using two sampling spots under different aerosol deposition rates.*"

All Figures: Please use larger font.

Response:

Lager font has been used in all figures.

Figure 2: Why do the ATN reach to above 400? This is never used with Aethalometers!

Response:

In this study, two Aethalometers were used to measure BC at different flows. The filter tape in these two Aethalometers was shifted automatically to expose a pristine spot on the filter at the same time every eight hours to ensure that the two sampling spots had a fixed starting and ending time of the sampling for internal comparison. When ambient BC was high, the measured ATN could rise to over 300 after 8 hr. And, the modified ATN could be high as >400.

Figure 4: Units are missing.

Response:

Units have been added in Fig. 4 and 5 (Fig. 5 and 6 in the revised manuscript).

Figure 5: Is this fig 4 with log scales? If so, why does the x-axis stop at ATN/Q=20 or 40? Make the x-axis identical for (a) and (b), preferably same as Fig. 4. See also comments to the manuscript text!

Response:

1. Yes.
2. The Fig. 5 (Fig. 6 in the revised manuscript) has been re-drawn with same scale.

Figure 6: Why is the y-axis stretched between 0.5 and 1? The values of (1-k.ATN) all lie between 0.8 and 1!

Response:

The Fig. 6 (Fig. 7 in the revised manuscript) has been re-drawn with a scale between 0.8 and 1.

Figure 7: The slopes need to be included on the figure. The axes maximums need to be reduced (potentially "breaking" them to include higher values) to show more detail at low BC concentrations. What about other wavelengths, especially 470 nm, since it was used to determine the Angstrom exponent?

Response:

The Fig. 7 (Fig. 8 in the revised manuscript) has been re-drawn with a suitable scale between 0 and 1000. The fitting results have been added in Fig. 7 (Fig. 8 in the revised manuscript). The figures for other wavelengths have been shown in supplement material. They had similar results with the Fig. 7 (Fig. 8 in the revised manuscript) for other channels.

Figure 8: The axes maximums need to be reduced to show more detail at low BC concentrations.

Response:

The Fig. 8 (Fig. 9 in the revised manuscript) has been re-drawn with a suitable scale between 0 and 1000.

While the drawbacks of the manuscript are by no means negligible and presentation should be improved significantly, the idea to combine two Aethalometers is interesting. I believe that the manuscript results could be better presented with appropriate major revisions as described above. These would make it suitable for publication in AMTD, most probably as a Technical Note.

Response:

Referee's remarks and comments have been carefully responded in the revised manuscript.