Responses to Review of Referee 2 (amt-2015-33)

We would like to acknowledge to the anonymous referee 2 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).

Filter-based absorption photometers suffers from several artifacts bounded to the presence of the filter matrix. One of them consists on a BC concentration depending on the loading of the filter. The paper presents a new method to correct for this loading effect by using simultaneously two Aethalometers with different flows leading to different filter loadings and allowing therefore to correct for the loading artifact. The presented method was applied on a one month and three months measurement campaign in winter and summer, respectively, in an urban environment.

General comments:

This paper presents a method to correct for the loading artifact of the "old versions" of the Aethalometer, since the firma MAGEE Scientific developed a new version AE33 using a similar method to correct for the loading effect (Drinovic et al., 2014). One disadvantage of this method is to need 2 Aethalometers to be applied. Climatology analysis need most of the time not the BC but the absorption coefficient as mentioned by the authors (p. 2862, Equ. 12). The main artifact of AE remains however the multiple scattering effect and the fact that the multiple scattering constant C (used in Equ. 12) depends on several factors not yet completely determined, such as the organic content (Lack et al., …), the aerosol size distribution (??) and the aerosol type (Collaud Coen et al., 2010). In this sense, the proposed correction method does not tackle the main artifact of AE's. This point has to be mentioned anywhere, so that the reader is aware that this method does not improve the major problem!

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.
   "*However, there is some uncertainly with this value due to different evaluation techniques. Collaud Coen et al. (2010) suggested that the C value ranged between 2.9 and 4.3 for the four different monitoring sites, indicating the C value was also dependent on aerosol properties (such as the amount of organic content).*"
3. The following statement has been addressed in the revised manuscript.
   "*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."

The last artifact is a dependence of the BC concentration on the aerosol scattering, which is overcome by the MAAP measuring also the backscattering of the embedded filters. The authors claim that their method also compensate for this artifact, what is however not proved in the paper. This artifact is however a much less important one.

Response:

The statement about the light scattering effect on measurement result has been modified and is no more emphasized.

Could you also please comment on the "best flow" to use to minimize the loading artifact? Would it be good to preload the filter to avoid the greatest loading effects at low ATN?

Response:

1. The loading effect at high flow rate is more serious than that at low flow rate. But, the light scattering effect on unload filter at low flow rate is more critical than that at high flow rate. Therefore, it is difficult to suggest a best flow. If the light scattering effect on unload filter is not an important issue, the lower flow rate is a best choice. However, negative values may be recorded at very low ambient concentration at lower flow rate.
2. It could be a good idea, but how to perform it in real world may be a big challenge.

Is it possible to apply this method with different versions of the AE, for example an AE31 and an AE16?

Response:

If they have the same sampling spot area and the same operation principle, they can be used to determine the BC concentration for a minimizing artifact using this proposed method.

Detailled comments:

- P. 2852, line 9: "in absence of sampling artifacts": the sampling is performed by the inlet and is not tested in this study. Did you mean "loading artifact" ?

Response:

1. "sampling artifacts" has been changed to "loading artifact".
2. "*In the absence of loading artifact, the ratio of ATN values measured by the two Aethalometers should be equal to the ratio of the sampling flow rates (or aerosol*

deposition rates) of these two Aethalometers."

- p. 2854, line 11: "Collaud Coen" instead of "Collaud Cone"

Response:

"Collaud Cone" has been changed to "Collaud Coen".

- Equ. 4: I suppose that you consider that t0=0 (as said in the text) and also ATN0=0.

Response:

The statement has been modified as following:

"*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*

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

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

- Have the dotted lines in Fig. 5 similar significance as in Fig. 4? Are Fig 4 and 5 done by fitting the results of the experiment? if yes, how good are the fits ?

Response:

Figs 4 and 5 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).

- P. 2857 line 18: "In this model, the correction factor k is assumed to be fixed for an ATN/Q value": Fig. 5 shows that this is not the case for very low ATN/Q values. These very low ATN/Q corresponds to very low ATN (ex: if Q=2, ATN=0.2 to obtain ATN/Q=0.1). Has it any signification to plot on such a large scale Fig. 4 and 5? Can you give the limits to obtain a k value that does not depend on ATN/Q?

Response:

1. Analytical results showed that the relationship between k (k_a in the revised manuscript) and ATN/Q for sampling flow rates of 6 and 2 L min^{-1} was similar, especially for long wavelengths. This result indicated that the assumption of the k_a value being fixed for an ATN/Q value in the proposed model was reasonable.
2. Analytical results indicated that the k_a value was negative at small ATN/Q values and it was a little different at a fixed ATN/Q, especially at short wavelengths in the winter season. The noise at very small ATN in the measurement could significantly contribute to the value of the parameter k_a. However, these extremely small ATN/Q values could be not important at actual sampling.

3. No, k_a is dependent on ATN/Q.

- P. 2860, last §: It would be interesting to have an idea about the difference between the k of Drinovec and the one of this study, perhaps to give mean, median and standard deviation for each case at high ATN/Q.

Response:

Correction factor k is not a constant value and it is dependent on ATN/Q. Moreover, the definitions of k for these two methods are different. Therefore, we can't provide the mean, median and standard deviation of k and compare them. Therefore, the correction results of BC were used to compare.

- P. 2861 line 3-5: did you calculate the relation between k and ATN/Q from equation 9? Or is figure 4 an experimental result? When you speak of "analytical results", did you mean that you can deduce it from equations? If yes, can you give the equation for k(ATN/Q)? If no, can you provide a statistical uncertainty from all the measurements? The same comments can be done for Fig. 5 and 6.

Response:

Figs 4-6 are analyzed from fitting equations based on all 5-min measurement data. The relation between k and ATN/Q was calculated from the relationship between measured ATN_{F6} and ATN_{F2}. 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).

- P. 2861 line 11: it seems from Fig. 5 that k is constantly decreasing with ATN/Q and it becomes not constant as stipulated in the text.

Response:

The statement has been changed as:

"*Finally, it was constantly decreasing with the increase of ATN/Q.*"

- P. 2861 line 15-17: as already said before, the fact that the effect of the light scattering in the filter matrix can be eliminated is not described/proved.

Response:

1. The statement about the light scattering effect on measurement result has been modified and is no more emphasized.
2. The statement has been changed 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."

- P.2862 line 7-9: the C constant depends not only on the filter material, but also on the aerosol properties such as the amount of organic content, the single scattering albedo,…

Response:

The following statement has been added in the revised manuscript.

"*However, there is some uncertainly with this value due to different evaluation techniques. Collaud Coen et al. (2010) suggested that the C value ranged between 2.9 and 4.3 for the four different monitoring sites, indicating the C value was also dependent on aerosol properties (such as the amount of organic content).*"

- P.2862, line 24-25: the proposed correction modify (increase) the Angström exponent, but, without have a reference, it is not possible to know if it is an improvement. It had to be noted than that the three given Angström exponents are in the confidence limits of each other, and that the confidence limits increase with the proposed correction. It seems then to me that the conclusions given in p. 2863 should be revised.

Response:

1. We just would like to express that the absorption Ångström exponent values computed using the ATN measured at the sampling flow rates of 6 and 2 L min^{-1} were relatively lower than those estimated from the modified ATN.
2. The confidence limit increase with the proposed correction was due to that the corrected absorption Ångström exponent values were significantly larger than that before corrected, particularly at high Ångström exponent value. After corrected, the absorption Ångström exponent values had a larger distribution range.
3. The statement has been modified as:

   "*Analytical results showed that the absorption Ångström exponent value could be raised from 1.01 ± 0.22 (estimated from measured* ATN_{F6}*) and 1.11 ± 0.33 (estimated from measured* ATN_{F2}*) to 1.17 ± 0.44 (estimated from modified* ATN_{F6} *or* ATN_{F2}*) in the summer season. In the winter season, the absorption Ångström exponent value could be increased from 1.01 ± 0.18 (estimated from measured* ATN_{F6}*) and 1.10 ± 0.20 (estimated from measured* ATN_{F2}*) to 1.15 ± 0.24 (estimated from modified* ATN_{F6} *or* ATN_{F2}*). The absorption Ångström exponent values computed using the ATN measured at the sampling flow rates of 6 and 2 L* min^{-1} *were relatively lower than those estimated from the modified ATN (*$p < 0.001$ *for both), indicating that the absorption Ångström exponent value could be significantly underestimated because of the filter loading effect.*"

- P. 2863, last § and p. 2864 first §: Is it possible to explain this effect by a different depth of deposition into the filter depending on the flow rate? Otherwise, do you have other possible explanation? Can the difference between winter and summer be due to the difference in aerosol concentrations?

Response:

The statement has been modified as:

"*For different sampling flow rates, the* $(1 - k_a \times ATN)$ *value differed significantly for the same ATN conditions because of different aerosol deposition rates, indicating that the aerosol deposition density on the filter and the depth of deposited aerosol into the filter could influence the extent of the aerosol loading effect on BC measurement results. Moreover, the aerosol loading effect on BC measurement results in the summer season was greater than that in the winter season by approximately 1%–6%, and it depended on the sampling flow rate. This difference between seasons could be because of differences in the composition, source, and age of aerosols, in addition to the different weather conditions.*"

- § 3.5: reading the text, one has the impression that there is great differences between both methods, the slopes presented in Fig. 8 are however always =1!

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 (Fig. 9 in the revised manuscript) are close to 1.

- Fig. 7 and 8: would it be better to decrease the scale range, even if there is some points at high BC

Response:

Fig. 7 and 8 (Fig. 8 and 9 in the revised manuscript) have been re-drawn with a suitable scale range.