Virkkula et al.: On the interpretation of the loading correction of the aethalometer, AMTD-2015-145

Replies to the reviewer's comments

First of all, the authors thank the reviewers for their evaluations, they helped improving the paper.

The text was corrected according to most of the suggestions of the reviewers. The largest change was the addition of one subsection describing possible implementation of the observed relationships into an algorithm. In that section also a probable source of uncertainty, the mixing state of aerosols is now mentioned. Some text regarding that is added also to the conclusions.

In the revised text major changes and additions are highlighted with yellow. Minor language corrections have not been indicated.

Below the **reviewers' comments are written with boldface fonts** and the replies with normal fonts, intended.

Detailed replies to Anonymous Referee #1

Page 7380, line 14: the instrument contains "mass attenuation coefficients", which is consistent with the notation of Eq. 3. The "uncorrected absorption coefficient" σ_0 is usually denoted as the attenuation coefficient". Using this notation might avoid unnecessary confusion on the part of the readers.

The reviewer is right. The term attenuation coefficient is now used but the symbols were not changed. The beginning of the section 2.3 was changed to

> "The aethalometer data were first used to calculate the uncorrected attenuation coefficients, here σ_0, by multiplying the original non-corrected BC mass concentration (BC_0 above) given by the aethalometer with the wavelength-dependent BC mass attenuation coefficient used by the instrument's software. Note that in several papers the symbol b_{ATN} has been used for the attenuation coefficient. In the present paper the symbol b is reserved for backscatter fraction, however, and the subscript of σ_0 is there to show that it was calculated from BC_0."

Figure 8: the discussion of this figure in section 3.4 is very interesting. The figure needs a legend with labels for Collaud Coen et al. (2010) and Arnott et al. (2005) algorithms.

A legend was added to the figure.

Detailed replies to Anonymous Referee #2

SPECIFIC COMMENTS

The only specific comment aims at a more clear recommendation of how the findings of this study should be used in the future analysis of aethalometer data. Actually, the interdependencies between the aethalometer compensation parameter k, the aerosol backscatter fraction, and aerosol single-scattering albedo are analysed, but their potential aimplementation in a new data analysis algorithm is missing. A separate short section on this topic would enhance the usefullness of the study significantly and may be included into Chapter 3.

We hesitate presenting any clear algorithm at this stage. The problem with implementing the data obtained from the compensation parameter into a data analysis algorithm is that there was not a proper absorption standard available. In addition, another source of uncertainty, the mixing state was not mentioned earlier. The mixing state may potentially also affect the compensation parameter which would make an implementation of an algorithm more difficult.

These thoughts were formulated into a new short section 3.6, as suggested by the reviewer:

3.6 Possible implementation into a data analysis algorithm

If the relationships between k, ω_0, and b were unambiguous aethalometer data could in principle be used in an algorithm for estimating ω_0 and b. However, as it was shown in the previous section, even after the classifications into ω_0 and b bins there was still a large range of compensation parameters that remained unexplained. A probable reason may have been rapidly varying BC mass concentrations, as mentioned several times above, but there are also other possible explanations.

An important factor missing in the present study is the mixing state of absorbing and scattering aerosols since there was no method available to measure it. It is likely that the same amount of absorbing aerosol such as BC yields different compensation parameters when it is internally mixed with scattering material and when these two are externally mixed. In these cases the penetration depths of BC particles into the filter would be different even if the overall backscatter fraction of the aerosol were the same. Therefore it is not likely there will be an unambiguous relationship between k, ω_0, and b. However, for internally mixed, typical aged BC aerosol, future research may prove out that the relationship is simple enough to be implemented into an algorithm.

MINOR ISSUES

1| In the manuscript the term BC mass concentration should be used instead of BC concentration.

Ok, it was changed all over.

2| The term "darkness of aerosol" or "dark aerosol" is not very precise. The authors may use a more specific term like, e.g., "light-absorbing aerosol".

We don't consider the expressions "dark aerosols" or "darkness of aerosols" bad at all, their use is not uncommon in scientific literature. Actually, they been used by several prominent authors, here are just four examples:

1) The famous paper presenting the CLAW hypothesis:
   Charlson et al. "Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate." *Nature* 326.6114 (1987): 655-661.
   "…effects on cloud albedo due to absorption of sunlight by **dark aerosol** particles …"

2) Arnott et al. "Towards Aerosol Light-Absorption Measurements with a 7-Wavelength Aethalometer: Evaluation with a Photoacoustic Instrument and 3-Wavelength Nephelometer" AST 39:17–29, 2005.
   "... For **dark aerosol** with a single-scattering albedo, $\omega \approx 0.3$, …"

3) Wilcox, E. M. "Direct and semi-direct radiative forcing of smoke aerosols over clouds." *Atmospheric Chemistry and Physics* 12.1 (2012): 139-149.
   "…Observations from Earth observing satellites indicate that **dark carbonaceous aerosols** that absorb solar radiation are widespread in the tropics and subtropics…."

4) Song, et al: "Black carbon at a roadside site in Beijing: Temporal variations and relationships with carbon monoxide and particle number size distribution" Atmos. Environ, 77, 213 – 221, 2013.
   "…The k factors in winter were larger, which could probably be explained by **darker aerosols**….

Two changes were done, however, regarding the word "darkness".
In the introduction of the AMTD paper P7376, L15-16 there is the sentence
   "It is definitely expected that the compensation parameter depends on the darkness of the particles,…

New words, single scattering albedo were added to clarify the term. In the revised introduction it now reads:
   "It is definitely expected that the compensation parameter depends on the darkness, i.e., the single-scattering albedo of the particles,

In the AMTD paper in section 3.5, P7390-7391, L2 it reads:
   "The darkest aerosol was observed when the backscatter fraction was the highest…

New clarifying words were added so that the sentence now reads:
   "The darkest aerosol, i.e., the aerosol with the lowest ω_0 was observed when the backscatter fraction was the highest …"

3| In all figures, the font size of axis labels should be checked, actually they are hardly readable.

The font sizes were increased in all figures and they were made clearer in other ways too, but the data shown are the same as in the discussion paper.

4| Page 7380, line 22: please explain the parameters on which the fraction of light scattering s depends.

The fraction s depends on several parameters described in different papers and describing them in detail. Actually repeating them would make this section very long. Instead of doing that a short text was added:

> " … is not any constant factor but also a wavelength-dependent function. For instance, if the algorithm of Arnott et al. (2005) is arranged as in (3) it is obvious that s depends on the absorption coefficient accumulated since the change of the filter spot and if the algorithm by Collaud Coen et al. (2010) is arranged as in (3) it can be seen that s is a function of the attenuation at any given time step, among other things. "

5|Please check carefully the use of English language: some examples are:

Page 7376, line 6, should be rephrased: "that the k value varies …"

done

Page 7376, line 8, should read "both at an urban and rural site"

done

Page 7377, line 3, should read "that site-related and seasonal …"

Done

Page 7385, line 12, the expression "close to similar" should be replaced by, e.g., either "close" or "similar".

done