L. Drinovec et al.: The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech. Discuss., 7, 10179–10220, 2014

REVIEW

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.

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.

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?

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

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

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

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

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 & σ 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)?

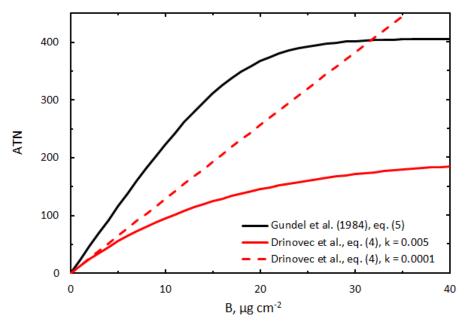


Figure 1. Comparison of Gundel et al. and Drinovec et al. parameterizations

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
$$BC = BC_{NC} \frac{1}{1 - kATN}$$

On the other hand, the sum of a geometric series

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

If |kATN| < 1, eq. (6) can be written $BC = BC_{NC}(1 + kATN + (kATN)^2 + ...)$

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.

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.

P10189, 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 ATN2/ATN1 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 (ATN2/ATN1) vs. ATN1".

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.

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 ATN₁) and spot 2 (ATN_{f2})? Again, explain very clearly, derive the formula and possibly use also graphical presentation. Don't leave space for speculations and misunderstandings.

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.

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.

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 $\sigma_{air} = 7.7 \text{ m}^2 \text{ g}^{-1}$, obtained initially by comparing optical and thermal measurements of filters loaded with refractory carbonaceous material (Gundel et al., 1984)."

What is σ_{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 m² 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 ± 1.2 m² 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.

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