A Simple Procedure for Correcting Loading Effects of Aethalometer Data

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ABSTRACT

A simple method for correcting for the loading effects of aethalometer data is presented. The formula $BC_{\text{CORRECTED}} =$ $(1 + k \cdot ATN) \cdot BC_{\text{NONCORRECTED}}$, where *ATN* is the attenuation and BC is black carbon, was used for correcting aethalometer data obtained from measurements at three different sites: a subway station in Helsinki, an urban background measurement station in Helsinki, and a rural station in Hyytiälä in central Finland. The BC data were compared with simultaneously measured aerosol volume concentrations (V). After the correction algorithm, the BC-to-V ratio remained relatively stable between consequent filter spots, which can be regarded as indirect evidence that the correction algorithm works. The k value calculated from the outdoor sites had a clear seasonal cycle that could be explained by darker aerosol in winter than in summer. When the contribution of BC to the total aerosol volume was high, the k factor was high and vice versa. In winter, the k values at all wavelengths were very close to that obtained from the subway station data. In summer, the k value was wavelength dependent and often negative. When the k value is negative, the noncorrected BC concentrations overestimated the true concentrations.

IMPLICATIONS

The aethalometer, a commonly used method for measuring BC concentrations in atmospheric aerosol, calculates the concentrations from the rate of change of light transmission through a filter; the faster the filter gets dark, the higher the BC concentrations are. The instrument software assumes that the relationship between the BC concentration and the rate of change of light transmission is linear; however, in most cases this is not true. The method presented in this work can easily be used for correcting this nonlinearity. The method also yields information on the contribution of BC to the aerosol volume.

INTRODUCTION

The origin of black carbon (BC) aerosol is in the incomplete combustion of fossil fuels and various types of biomass burning, ranging from small-scale residential wood combustion to large forest fires. BC aerosols have significant adverse health effects and they also play an important role in climate forcing because they are the most important contributor to light absorption by aerosols. BC is therefore measured in urban, rural, and background areas worldwide.

The aethalometer¹ is probably the most common method for measuring BC concentrations. In the method, air is drawn through a filter and the decrease of light transmission through the sampling area A is measured. Decreasing transmission leads to increasing attenuation, $ATN \equiv -\ln(I/I_0)$, where I_0 is the light intensity of the incoming light and *I* is the light intensity after passing the filter. In the aethalometer it is assumed that the *ATN* increase is only because of light absorption by BC accumulating on the filter, and BC concentration is therefore calculated from the rate of change of attenuation:

$$BC = \frac{\sigma_{\rm abs}}{\alpha_{\rm abs}} = \frac{1}{\alpha_{\rm abs}} \frac{A}{Q} \frac{\Delta ATN}{\Delta t}$$
(1)

where σ_{abs} is the particle absorption coefficient, α_{abs} the mass absorption cross section of BC, and Q is the airflow rate through the filter. It is well known, however, that the relationship between *ATN* change and BC concentration is not linear.^{2–6} There are several reasons for this, including that both scattering and absorbing particles collected on the filter alter the internal reflection of the filter in a way that changes the absorption of the aerosol/filter combination.^{3,7–9} There are two main consequences: (1) as the filter gets darker, that is, as *ATN* increases the measured BC concentration gets underestimated; and (2) scattering aerosol gets interpreted as BC. The first of these is the more important one. These effects may be taken into

account using empirical correction functions, such as that derived by Weingartner et al.⁴ Arnott et al.⁶ derived a model-based algorithm for the correction of aethalometer data. Both of these correction methods take both aerosol scattering and absorption coefficients into account. In the new instrument, the Multi-Angle Absorption Photometer (MAAP), these effects are taken into account already in the design and an internal algorithm of the instrument.^{10,11}

In this work an alternative correction algorithm is presented for the aethalometer data. The procedure does not take scattering into account because many organizations use the aethalometer in the field without any instrument that measures scattering and such a procedure is needed. The principle of the algorithm is presented and then applied to three different datasets: measurements at a Helsinki city subway station; at an outdoor urban air measurement station in Helsinki; and at Hyytiälä, a rural station in a forest in southwestern central Finland.

ALGORITHM

The operation principle of the aethalometer is very close to that of the Particle Soot Absorption Photometer (PSAP)^{3,12} so formulas developed for the PSAP data are used here. Virkkula et al.¹² derived an empirical correction formula for the PSAP data

$$\sigma_{\text{abs}} \text{ (corrected)} = (k_0 + k_1 \ln(I/I_0))\sigma_0 - s\sigma_{\text{SP}}$$
(2)

where k_0 , k_1 , and *s* are empirically derived constants; σ_0 is the noncorrected absorption coefficient, defined essentially the same way as σ_{abs} in eq 1; and σ_{SP} is the particle scattering coefficient. Using the definition of *ATN* above, eq 2 becomes

$$\sigma_{\rm abs} \text{ (corrected)} = (k_0 - k_1 A T N))\sigma_0 - s\sigma_{\rm SP}$$
(3)

It is assumed here that the correction function for the aethalometer data is of the same form. In the aethalometer data files *ATN* is presented as $100 \times (-\ln(I/I_0))$, so for simplicity this *ATN* will be used in the algorithm. It is also assumed here that the raw *BC* given by the aethalometer is correct when the filter is clean, that is, when ATN = 0. Several aethalometer users do not have a nephelometer in use and they have no information on σ_{SP} , so the algorithm presented here does not have it either. It follows that for the correction algorithm $k_0 = 1$ and s = 0, there is only one constant to be found. The corrected absorption coefficient then becomes

$$\sigma_{abs} \text{ (corrected)} = (1 + k \cdot ATN) \sigma_{abs} \text{ (noncorrected)}$$
(4)

and the corrected BC concentration is calculated from

$$BC_{\text{CORRECTED}} = \frac{\sigma_{\text{abs}} \text{ (corrected)}}{\alpha_{\text{abs}}} = (1 + k \cdot ATN)BC_0$$
(5)

where BC_0 is the noncorrected BC concentration given by the aethalometer. This notation will be used in the text below. Probably the most used operational mode of the aethalometer is such that it collects the sample on a filter tape that moves forward when *ATN* through the spot has reached a preset limit—in the data to be discussed below the spot change took place when *ATN* was approximately 75%—and starts measuring the next spot. A value for the factor *k* in eq 5 is calculated for each filter spot so that the data become continuous, that is,

$$BC_{\text{CORRECTED}}(t_{i,\text{last}}) = BC_{\text{CORRECTED}}(t_{i+1,\text{first}})$$
(6)

where $t_{i,last}$ is the time of the last measurement data for filter spot *i*, and $t_{i+1,first}$ is the time of the first measurement data for the next filter spot. Applying eq 5 in eq 6, we obtain the formula for calculating the factor *k* for the filter spot *i*:

$$k_{i} = \frac{BC_{0}(t_{i+1,first}) - BC_{0}(t_{i,last})}{ATN(t_{i,last}) \cdot BC_{0}(t_{i,last}) - ATN(t_{i+1,first}) \cdot BC_{0}(t_{i+1,first})}$$
(7)

This is the general form for calculating k_i . In typical atmospheric conditions this can be somewhat simplified. Right after the filter spot has been changed the first $ATN \approx 0$ so eq 7 becomes

$$k_{\rm i} \approx \frac{1}{ATN(t_{\rm i,last})} \left(\frac{BC_0(t_{\rm i+1,first})}{BC_0(t_{\rm i,last})} - 1 \right) \tag{8}$$

The obtained factor ki is then used for correcting all data obtained for filter spot *i* according to eq 5. In practice, *ATN* is not exactly 0 even in the first data line unless the BC concentration equals zero. The BC data have also noise that is not due to the effect discussed here, i.e., instrumental noise or true variation in BC concentrations. Therefore, in practice the *k* factors were calculated from eq 8 using the average BC_0 of the last three data of filter spot *i* and the first three data of filter spot i + 1 for the measurements that had a 1-min time resolution and the average of last two data of filter spot *i* and the first two data of filter spot i + 1 for the measurements that had a 5-min time resolution.

In case the aerosol contains significant amounts of BC, i.e., in exhaust gas or smoke measurements not presented in this paper, the *ATN* of the first measurement data of the new filter spot is already clearly higher than zero. In this case the assumption that the first *ATN* \approx 0, which was used to obtain eq 8 is not valid and eq 7 has to be used.

MEASUREMENTS

Three datasets are used for testing the algorithm. First, the measurements in a Helsinki city subway station are analyzed in more detail than the other data. The reason is that these data are the simplest and the time resolution is the highest so they are the best data for presenting the

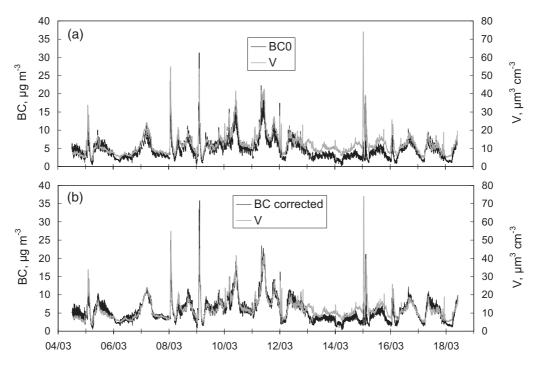


Figure 1. (a) Noncorrected (*BC*₀) and (b) corrected BC concentrations and aerosol volume concentrations during the whole measurement campaign in the underground subway tunnel.

strengths and weaknesses of the method. Next, the algorithm is applied to two different types of atmospheric aerosols: urban and rural.

The subway measurements were part of a large campaign to evaluate the exposure to particulate matter in the subway system of Helsinki. The main results of the campaign were presented earlier by Aarnio et al.¹³ In the campaign, measurements were carried out at two surface stations: one ground-level station and in subway cars. In this paper, the data of the measurements carried out at the underground subway station of Sörnäinen in the center of Helsinki from March 4–18, 2004, are used.

The measurement setup was described in detail by Aarnio et al.¹³ so here only a short summary is presented. The instruments were installed approximately 1 km from the tunnel entrance, on the roof of a maintenance room, approximately 4 m above the platform level. The instruments took their sample air through a fine particulate matter PM_{2.5} inlet. The measurements included PM_{2.5} mass monitoring, filter sampling for subsequent chemical analyses, particle number size distribution measurements, and BC measurements. The BC data were obtained from a one-wavelength aethalometer AE-16, manufactured by Magee Scientific, Berkeley, CA, that uses an light-emitting diode at $\lambda = 880$ nm. Particle number size distributions in the diameter size range 10-500 nm were measured with a differential mobility particle sizer (DMPS). The time resolution of the BC and DMPS measurements were 1 and 5 min, respectively. The time between the last and first data points of two consecutive aethalometer filter spots was 6 min. The aethalometer sample flow rate was 4 liters per minute (LPM). In this paper the DMPS data are used for calculating aerosol volume concentration, V, in the respective size range.

The same aethalometer was used for measuring outdoor aerosol at an urban background station, SMEAR III (System for Measuring Forest Ecosystem-Atmosphere Relationships III) (www.atm.helsinki.fi/SMEAR/), in Kumpula, Helsinki (60°20'N, 24°97'E, 26 m above sea level) in March-September 2006.14 The aethalometer inlet removed particles larger than 2.5 µm. The site is located in the campus area of the University of Helsinki at a distance of 5 km northeast from the center of Helsinki. The most important local source of BC at this site is a major road at a distance of 200 m to the east. The time resolution of the BC data was 5 min, and the time between the last and first data points of two consecutive filter spots was 15 min. Particle size distributions in the size range 3–950 nm were measured with a Twin-DMPS system with a 10-min time resolution. In this paper only the integrated volume size distributions are used.

A seven-wavelength aethalometer, model AE-31, operating at wavelengths of 370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm, and 990 nm, was used at the Hyytiälä boreal forest ecosystem measurement station, SMEAR II^{15,16}, from December 2004 through September 2006. The station is located at Hyytiälä, in southern Finland (61°51′N, 24°17′E, 170 m above sea level). The time resolution of the BC data is 5 min, and the time between the last and first data points of two consecutive filter spots is 15 min. The aethalometer inlet did not have a specific cutoff diameter. Particle size distributions in the size range 3–500 nm were measured with a Twin-DMPS system with a 10-min time resolution.¹⁶ In this paper only the integrated volume size distributions are used.

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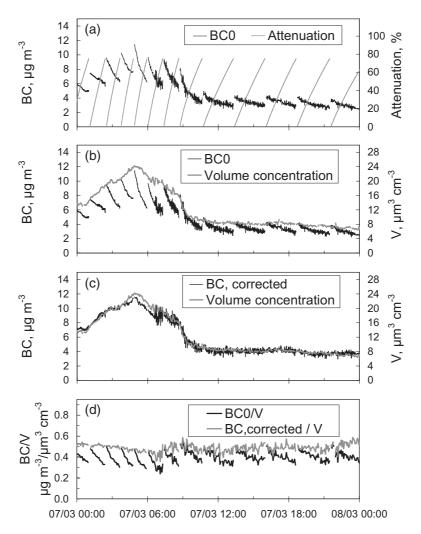


Figure 2. Subway tunnel aethalometer and DMPS data on March 7, 2004. (a) Noncorrected BC concentrations and aerosol volume concentrations, (b) noncorrected BC concentrations (BC_0) and attenuation, (c) corrected BC concentrations and aerosol volume concentrations, and (d) ratio of noncorrected and corrected BC concentrations to aerosol volume concentrations.

RESULTS AND DISCUSSION Subway Station Data

The time series of the noncorrected and corrected BC concentration and aerosol volume concentration, V, measured during the whole campaign are presented in Figure 1. It is obvious even before any corrections that BC and volume concentrations follow each other well and that a significant fraction of the particles consists of BC. The exact fraction cannot be calculated from these data because the DMPS measured particles with $D_{\rm p} < 500$ nm and the aethalometer was connected to a $PM_{2.5}$ inlet. However, the BC concentrations correlated very weakly with $PM_{2.5}$ as the time series presented by Aarnio et al.¹³ showed. The very good correlation with V and the bad correlation with PM_{2.5} indirectly suggest that BC was mainly in the particle size range $D_{\rm p} < 500$ nm. After March 12, BC and V did not correlate quite as well. The most probable explanation for this is that the inlet used by the aethalometer was partially clogged. This was observed when the campaign was over, the inlet was opened, and a pile of impacted particulate matter was

observed. This will not be pursued further, however, because for this paper the most significant information in Figure 1 is that the noncorrected BC concentrations have clear discontinuities that are not present in the V data, and that the correction algorithm presented above removes most of this discrepancy. On March 7, the phenomenon was very clear so this day is analyzed in more detail.

The time series of the noncorrected and corrected BC concentrations, *ATN*, and *V*, measured on March 7 are presented in Figure 2. The noise in BC concentration after 6:00 a.m. is due to the start of the subway ventilation system, the regular train traffic, and the resulting air turbulence in the tunnel. Panel (a) clearly shows how the noncorrected BC concentration starts decreasing immediately after each filter spot change. Panel (b) shows that the BC concentrations at the start of each filter spot well follow the aerosol volume concentration but not later when *ATN* has increased. Panel (c) shows that after using the above algorithm the corrected BC follows *V* very closely over the whole period, including periods with

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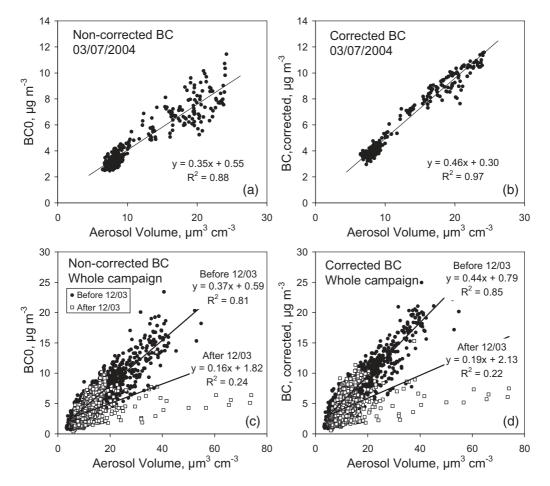


Figure 3. (a) Noncorrected and (b) corrected BC concentration vs. aerosol volume concentration at the subway station on March 7, 2004. (c) Noncorrected and (d) corrected BC concentration vs. aerosol volume concentration during the whole campaign.

higher *ATN*, and that the BC fraction of *V* does not vary significantly. The *BC*-to-*V* ratio plotted in (d) varied significantly for each spot when the noncorrected BC was used in the ratio but not when the corrected BC was used. A scatter plot of BC versus *V* for that day (Figure 3) shows that the squared correlation coefficient R^2 was significantly improved, from 0.88 to 0.97, after applying the correction algorithm. For the data taken before March 12 there was some improvement of R^2 (Figure 3) but after that the correlation was weak. An explanation for this is that after March 12 the inlet was probably clogged, as explained above.

Another reason that the correlation of BC and V was not improved as much for the whole dataset as for the 1-day period, March 7, is that there were periods when concentrations varied rapidly and the algorithm for deriving k did not work well. A good example of this is the early morning of March 8 (Figure 4). At 1:15 a.m., a diesel-powered service train passed the station and both V and BC concentrations rapidly increased approximately 10-fold. The usual discontinuities in the BC data are obvious in Figure 4a. At 1:41 a.m. the aethalometer changed the filter spot and the measurement continued at 1:47 a.m. In Figure 4b the BC data were corrected with eq 5 using two values for the factor k: (1) the k obtained from the usual algorithm that

combines the data so that it matches the last measurement data point of the spot that was finished at 1:41 a.m. with the first measurement data point of the spot that was started at 1:47 a.m. ($f(k_i)$) × BC₀), and (2) the k that was averaged over all k values from the whole measurement period ($f(k_{ave}) \times BC_0$). In case 1, the constant BC concentration over the spot change is unrealistic, on the basis of the observed smooth decrease of Vat a rate of approximately 2.4%/min after the maximum V was reached. It is also most probable that the BC concentrations decreased smoothly after the maximum was reached. When the correction was done using the average k the rate of the BC decrease was approximately 2.9%/min, close to that of the rate of V decrease, suggesting that during rapidly changing concentrations the use of average k gives a more reasonable correction than the individual k values. The BC-to-V ratio varied much more during this episode than normally, first decreasing to approximately 0.2 and then rising to >0.6, both with and without the BC correction (Figure 4c). The data are not sufficient for explaining the reason for this variation. However, the main message of this figure is that the BC-to-V ratio is not in all cases a good criterion for evaluating the procedure.

In the whole period the average standard deviation of k was 0.0051 \pm 0.0023. Most of the time it did not vary

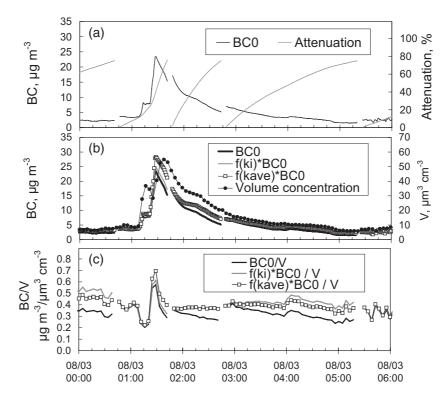


Figure 4. Example of spot changes where the individual *k* gives worse result than the average *k*. (a) Uncorrected BC data (BC₀) and attenuation, (b) BC₀ and BC data corrected with two different correction functions; $f(k_i)$: correction function using the individual *k* for each spot; $f(k_{ave})$: correction function using the average *k* for all spots; (c) BC-to-volume ratio using the uncorrected and corrected BC concentration. Subway tunnel data.

much from spot to spot but there were some clear deviations (Figure 5). Of the *k* values, 2.5% were less than 0. The negative values were all associated with rapidly decreasing concentrations. During rapidly increasing concentrations the *k* factor became greater than 0.01. In these extreme values the forcing of the concentrations to be the same before and after the spot change leads very probably to erroneous concentrations. The factor *k* varied more at low BC concentrations but there was no clear differences in the average values at low and high BC concentrations (Figure 6).

Outdoor Station Data

The algorithm was applied to the data from SMEAR II (Hyytiälä) and SMEAR III (Helsinki). The factor k was calculated for each spot change but to remove the highest and lowest values associated with rapid concentration changes, a running median of eight consecutive spot changes was used. The long time series from the Hyytiälä station reveals that there is a clear and strong seasonal variation of k (Figure 7). It can be observed also in the urban station data although the time series is shorter.

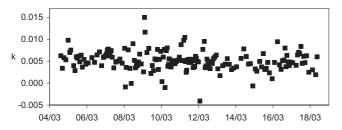


Figure 5. The factor k for the subway tunnel aethalometer data.

The interesting and important point in the time series is that the k values become clearly negative in summer. When the subway station data were discussed above, it was stated that the negative values were associated with rapidly decreasing concentrations. However, in the outdoor aerosol data even the monthly average k values become negative in summer (Figure 7), so this is indicating something else. From eq 5 it is obvious that negative k implies that the noncorrected BC overestimate the BC concentrations. When plotted against *BC*-to-*V* ratios, it is obvious that the k factor depends clearly on the contribution of BC to the total aerosol volume, so that at low BC-to-V ratios k is negative and at high ratios it is positive (Figure 8). This can be explained by the response of the aethalometer to scattering aerosol, as discussed by Weingartner et al.⁴ and Arnott et al.⁶ In summer, the contribution of BC to aerosol mass is smaller than in winter, which means that the contribution of scattering aerosol to total

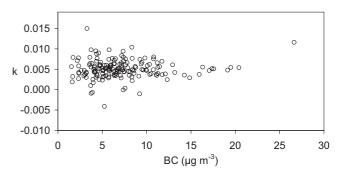


Figure 6. The factor k vs. BC at ATN = 0 for the subway tunnel aethalometer data.

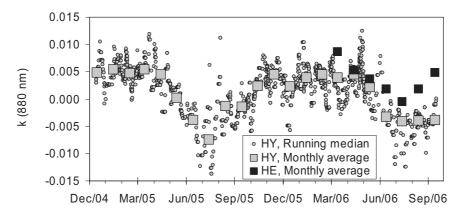


Figure 7. The factor k at $\lambda = 880$ nm at a rural station (HY, Hyytiälä) and at an urban station (HE, Helsinki). For the Hyytiälä data the running median of 8 consecutive spot changes and the monthly average are presented, and for the Helsinki data the monthly averages are presented.

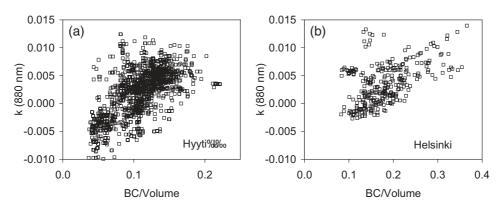


Figure 8. The factor k at $\lambda = 880$ nm as a function of the fraction of *BC*-to-*V* concentration ratio at (a) Hyytiälä and (b) Helsinki.

aerosol mass is higher in summer at these sites. In winter, the concentration and the contribution of BC to aerosol mass concentration is in general higher than in summer. The reasons include higher energy production by various combustion processes in winter and higher contribution of biogenic aerosol in summer.

In winter, the *k* factor at Hyytiälä did not depend significantly on wavelength (Figure 9). It is worth noticing that in winter the average standard deviation of *k* of all wavelengths was 0.0053 ± 0.0021 . This is practically

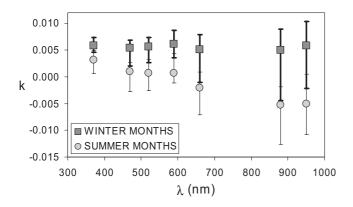


Figure 9. The median factor k in winter (November–April) and summer (June–August) at the Hyytiälä station. The error bars contain 95% of the data.

the same as the one derived from the subway station data above and the average *k* from the urban station in winter (Table 1). Contrary to winter data, *k* varied strongly with wavelength in summer (Figure 9). The average *k* at $\lambda =$ 370 nm was 0.0059 ± 0.0007 in winter and only slightly lower (0.0030 ± 0.0011) in summer, whereas for the $\lambda =$ 950 nm data, the respective values were 0.0055 ± 0.0031 and -0.0051 ± 0.0028. The explanation for this wavelength-dependent seasonal variation requires further analyses of aerosol chemical composition and optical properties, which is out of the scope of the present paper.

CONCLUSIONS

Most aethalometer users have observed that the apparent BC concentrations rise after the filter spot has been changed. In this work it is assumed that the first values after the spot change are closest to the real concentrations, i.e., the values measured when the filter is cleanest and $ATN \approx 0$. The formula $BC_{\text{CORRECTED}} = (1 + k \cdot ATN) \cdot BC_{\text{NONCORRECTED}}$ was used for correcting aethalometer data and a value for the factor k was calculated for each filter spot so that the data became continuous over the spot change. There was no independent absorption standard in use so we cannot really prove that the algorithm makes a true correction. However, it is a realistic assumption that the BC fraction of the total aerosol volume concentration V does not change significantly after

Table 1. The average (\pm standard deviation) factor *k* at the three measurement sites.

	370 nm	470 nm	521 nm	590 nm	660 nm	880 nm	950 nm
HY, WI	0.0059 ± 0.0007	0.0052 ± 0.0012	0.0054 ± 0.0012	0.0061 ± 0.0013	0.0047 ± 0.0021	0.0043 ± 0.0032	0.0055 ± 0.0031
hy, su he, wi	0.0030 ± 0.0011	0.0006 ± 0.0015	0.0006 ± 0.0015	0.0008 ± 0.0013	-0.0024 ± 0.0020	$\begin{array}{c} -0.0056 \pm 0.0030 \\ 0.0054 \pm 0.0018 \end{array}$	-0.0051 ± 0.0028
he, su Metro						$\begin{array}{l} 0.0007 \pm 0.0031 \\ 0.0051 \pm 0.0023 \end{array}$	

Notes: HY = Hyytiälä; HE = Helsinki; METRO = the subway station. The Hyytiälä and Helsinki data were averaged over the winter (WI = November-April) and summer (SU = June-August) months.

each filter spot change. Thus, the fact that after the correction algorithm the *BC*-to-*V* ratio remained relatively stable between consequent filter spots can be regarded as indirect evidence that the correction algorithm works.

The original idea for the method was obtained from the similarities of the PSAP and the aethalometer; therefore, it is worth comparing the average factor *k* with the corresponding factor for the PSAP. Virkkula et al.¹² obtained the average correction function $f(I/I_0) \approx 0.31$ – 0.53 $\ln(I/I_0)$ for laboratory-produced black aerosol for a 3-wavelength PSAP. Noting that in the aethalometer data ATN is presented as $100 \times (-\ln(I/I_0))$ the above PSAP function becomes f(ATN) = 0.31 + 0.0053 ATN. The factor 0.0053 that determines the dependence on ATN is essentially the same as the *k* obtained here from the metro tunnel data and from the average winter data from both the urban and the rural stations. Taking into account that the instruments use different filter materials this is an interesting observation emphasizing the similarities of methods that derive absorption coefficient from transmittance changes.

The main problem in the application of the algorithm is that the aethalometer has a calibration period after each filter tape move and it draws air through the filter spot even before measurement data are saved. This has two effects for the application of the algorithm. First, at high BC concentrations the spot gets dark even before the measurements start and there are no data with $ATN \approx 0$. This applies especially to the multiwavelength aethalometers because during the calibration period there is typically a break of approximately 20 min between the last measurement before the spot change and the first measurement after the change. Another problem in the algorithm is that it is assumed that the concentration does not vary much during the calibration period so it only works when the BC concentration remains reasonably stable. Fast concentration variations lead to unrealistic k values. For instance, if the true BC concentration is clearly higher just before the spot change, then after it the derived k becomes negative. During rapidly changing concentrations the use of average k gives a more reasonable correction than the individual *k* values.

More significant variations in k were observed in a long time series from a rural station, even though the data were cleaned of the values associated with rapidly changing concentrations. A clear seasonal variation of k was observed so that in winter the values were positive and very close to those obtained from the measurements in the metro tunnel and in summer they were lower and even negative for some wavelengths. Negative k values imply that the noncorrected BC values overestimate the true BC values.

Some of the variation of k could probably be explained by variations in the size distributions of the light absorbing particles. The mass absorption efficiency is a function of particle size and the BC data were all calculated with constant mass absorption efficiencies. It is probable that this is not the most significant explanation, though. For the PSAP it was observed that the factor k_1 varied with the single-scattering albedo (ω_0) = scattering/extinction) of the aerosol.¹² It is most probable that this is the case also for the aethalometer, so variation in ω_0 is a very likely explanation for the seasonal variation of the *k* factor obtained in this work. This is in agreement with the observed positive relationship between the k factor and BC-to-V ratio. At high *BC*-to-*V* ratios ω_0 is lower than at low ratios. Therefore a very interesting consequence of the method is that the aethalometer has the potential to yield information on not only BC concentrations but probably also on single-scattering albedo. To assess how quantitative the relationship between k and ω_0 can be, measurements should be conducted together with a nephelometer and some standard absorption method, such as a photoacoustic instrument or an extinction cell.

Even though the method presented here reduces the saw-like variations of BC concentrations over the filter spot changes, an important factor is missing: subtraction of the scattering coefficient, if it is available. The way this can be incorporated in the algorithm in a simple way must be investigated. The method in this paper was designed for correcting aethalometer data in case scattering coefficient data are not available, like in the measurements analyzed here, so this effect was not discussed further. However, the work required for the evaluation of the method includes a quantitative comparison with previously presented methods that do take this effect into account.^{4,6}

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REFERENCES

- Hansen, A.D.A.; Rosen, H.; Novakov, T. The Aethalometer—an Instrument for the Real-Time Measurement of Optical Absorption by Aerosol Particles. *Sci. Tot. Environ.* **1982**, *36*, 191.
- Reid, J.S.; Hobbs, P.V.; Liousse, C.; Vanderlei Martins, J.; Weiss, R.E.; Eck, T.F. Comparisons of Techniques for Measuring Shortwave Absorption and Black Carbon Content of Aerosols from Biomass Burning in Brazil; J. Geophys. Res. **1998**, 103, 32031-32040.
- Bond, T.C.; Anderson, T.L.; Campbell, D. Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols; *Aerosol Sci. Technol.* **1999**, *30*, 582-600.
- Weingartner, E.; Saathoff, H.; Schnaiter, M.; Streit, N.; Bitnar, B.; Baltensperger, U. Absorption of Light by Soot Particles: Determination of the Absorption Coefficient by Means of Aethalometers; *J. Aerosol Sci.* 2003, *34*, 1445-1463.
- Arnott, W.P.; Moosmüller, H.; Sheridan, P.J.; Ogren, J. A.; Raspet, R.; Slaton, W.V.; Hand, J.L.; Kreidenweis, S.M.; Collett, J.L., Jr. Photoacoustic and Filter-Based Ambient Aerosol Light Absorption Measurements: Instrument Comparisons and the Role of Relative Humidity; *J. Geophys. Res.* 2003, *108*, 4034, doi:10.1029/2002JD002165.
- Arnott, W.P.; Hamasha, K.; Moosmüller, H.; Sheridan, P.J.; Ogren, J.A. Towards Aerosol Light Absorption Measurements with a 7-Wavelength Aethalometer: Evaluation with a Photoacoustic Instrument and 3-Wavelength Nephelometer; *Aerosol Sci. Technol.* 2005, 39, 17-29.
- Clarke, A.D. Effects of Filter Internal Reflection Coefficient of Light Absorption Measurements Made Using the Integrating Plate Method; *Appl. Opt.* **1982**, *21*, 3021-3031.
- Clarke, A.D.; Noone, K.J.; Heintzenberg, J.; Warren, S.G.; Covert, D.S. Aerosol Light Absorption Measurement Techniques: Analysis and Intercomparisons; *Atmos. Environ.* **1987**, *21*, 1455-1465.
- Horvath, H. Experimental Calibration for Light Absorption Measurements Using the Integrating Plate Method—Summary of the Data; *J. Aerosol Sci.* 1997, 28, 1149-1161.
- Petzold, A.; Schönlinner, M. Multi-Angle Absorption Photometry—a New Method for the Measurement of Aerosol Light Absorption and Atmospheric Black Carbon; J. Aerosol Sci. 2004, 35, 421-441.
- Petzold, A.; Schloesser, H.; Sheridan, P.J.; Arnott, W.P.; Ogren, J.A.; Virkkula, A. Evaluation of Multi-Angle Absorption Photometry for Measuring Aerosol Light Absorption; *Aerosol Sci. Technol.* 2005, 39, 40-51.
- Virkkula, A.; Ahlquist, N.C.; Covert, D.S.; Arnott, W.P.; Sheridan, P.J.; Quinn, P.K.; Coffman, D.J. Modification, Calibration and a Field Test of an Instrument for Measuring Light Absorption by Particles; *Aerosol Sci. Technol.* **2005**, *39*, 68-83.

- Aarnio, P.; Yli-Tuomi, T.; Kousa, A.; Mäkelä, T.; Hirsikko, A.; Hämeri, K.; Räisänen, M.; Hillamo, R.; Koskentalo, T.; Jantunen, M. The Concentrations and Composition of and Exposure to Fine Particles (PM_{2.5}) in the Helsinki Subway System; *Atmos. Environ.* 2005, 39, 5059-5066.
- 14. Saarikoski, S.; Sillanpää, M.; Timonen, H.; Saarnio, K.; Teinilä, K.; Sofiev, M.; Karppinen, A.; Kukkonen, J.; Hillamo, R. Chemical Composition of Aerosols during a Major Biomass Burning Episode over Northern Europe in Spring 2006: Experimental and Modelling Assessments; Atmos. Environ. 2007, 41, 3577-3589.
- Hari, P.; Kulmala, M. A Station for Measuring Ecosystem-Atmosphere Relations (SMEAR ii); *Boreal Environ. Res.* 2005, 10, 315-322.
- Dal Maso, M.; Kulmala, M.; Riipinen, I.; Wagner, R.; Hussein, T.; Aalto, P.P.; Lehtinen, K.E.J. Formation and Growth of Fresh Atmospheric Aerosols: Eight Years of Aerosol Size Distribution Data from SMEAR II, Hyytiälä, Finland; *Boreal Environ. Res.* 2005, 10, 323-336.

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