



Supplement of

The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation

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24 Filter loading effect for different measurement sites and seasons

The filter loading effect is determined by the *BC*(*ATN*) method for measurement campaigns conducted at different types of the measurement site (roadside, urban background, rural background, regional background) in Klagenfurt (Austria), Anaheim (USA), Payerne (Switzerland), Sonnblick (Austria), Kathmandu (Nepal). Uncompensated *BC* data as measured by the AE33 with the detector under spot S1 was fitted using a linear function in the *ATN* range from 4.4 to 45. The filter loading effect is determined as the relative slope *RS* (Eq. 3).

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Figure S1. *BC*(*ATN*) analysis of the filter loading effect for the campaign in Klagenfurt (Austria) from 1-12 March 2012. The Klagenfurt station is located at Völkermarkter Strasse (46°37'32.1"N 14°19'05.7"E, 446 m ASL). The station is located in the middle of a crossroad and is strongly influenced by local traffic. During winter the site is strongly influenced by biomass burning from residential heating. Average BC data for each ATN bin (1 ATN unit wide) and the linear fit of the raw data are presented.





Figure S2. *BC*(*ATN*) analysis of the filter loading effect for the campaign in Anaheim (California,
USA) from 1-9 July 2012. The Anaheim station is located in an urban environment (33° 49' 50" N,
117° 56' 18"W, 39 m ASL). Average BC data for each ATN bin (1 ATN unit wide) and the linear fit of
the raw data are presented.



Figure S3. *BC*(*ATN*) analysis of the filter loading effect for the campaign in Payerne (Switzerland)
from 15 June – 5 July 2012. The Payerne aerological station (46.82 ° N, 6.95 ° E, 491 m ASL) is a rural
background air quality monitoring station located in Southwestern Switzerland, on the Swiss
plateau between the Jura and the Alps. It lies about 1 km south-east of the small rural town of
Payerne. The site is surrounded by agricultural fields (grassland and crops), forests and small
villages. Average BC data for each ATN bin (1 ATN unit wide) and the linear fit of the raw data are
presented.





Figure S4. *BC*(*ATN*) analysis of the filter loading effect for the campaign in Sonnblick (Austria) from 1 July to 31 August 2013. The Sonnblick observatory is a background station located in the high alpine environment (47°03'15"N, 12°57'27"E, 3106 m ASL). Measurements at this location allow the determination of the composition of the mid-troposphere, frequently reaching into the free troposphere. Average BC data for each ATN bin (1 ATN unit wide) and the linear fit of the raw data are presented.







73 Table S1. Average filter loading effect as determined with the *BC*(A*TN*) analysis for different

74 measurement sites and seasons.

Location	BC(ATN) relative slope RS	Site description	Season
Klagenfurt (Austria)	- 0.0069 +/- 0.0007	Roadside	Winter
Anaheim (USA)	- 0.0033 +/- 0.0003	Urban	Summer
Payerne (Switzerland)	- 0.0013 +/- 0.0004	Rural background	Summer
Sonnblick (Austria)	- 0.0017 +/- 0.0002	Regional background	Summer
Kathmandu (Nepal)	- 0.0047 +/- 0.0003	Urban	Winter

Channel	Wavelength (nm)	$\sigma_{air}(m^2g^{-1})$
1	370	18.47
2	470	14.54
3	520	13.14
4	590	11.58
5	660	10.35
6	880	7.77
7	950	7.19

80 Table S2. Mass absorption cross-section values used in the AE33.

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The mass absorption cross-section values σ_{air} denote the mass absorption cross-section of aerosols freely suspended in the atmosphere. The values used in the Aethalometer assume an inverse dependence on the wavelength – that is, a "black" sample with an Ångström exponent equal to unity (Moosmüller et al., 2011).

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The relationship between the absorption and mass was determined by optical measurements of transmission and thermal measurements of samples, where the non-refractory constituents of the carbonaceous sample were removed (Gundel et al., 1981; Gundel et al., 1984). The pollutants in ambient air and combustion source exhausts were sampled on quartz fiber filters and analyzed. The non-destructive optical measurement was the laser transmission method carried out at room temperature. The thermal method employed was the evolved gas analysis (EGA) in oxygen (Malissa et al., 1976). EGA was performed on the non-treated filters and on the filters sequentially extracted

95 by benzene and a mixture of methanol-chloroform. The most refractory peak in the extracted filter 96 thermograms was used to quantify black carbon. The mass attenuation cross section for the laser was thus obtained: $\sigma = (23.9 \pm 2.0) \text{ m}^2\text{g}^{-1}$. The high value is typical for the attenuation measurement 97 setups using collimated laser beams (Gundel et al., 1984). In the first Aethalometer, using an 98 99 incandescent lamp and a green band-pass filter (Hansen et al., 1984), the mass attenuation cross section was determined to be 10 m²g⁻¹ at 530 nm. In the ensuing commercial embodiment of the 100 101 Aethalometer, again using an incandescent lamp without any filters, the mass attenuation cross 102 section was determined to be 19 m²g⁻¹ (Babich et al., 2000). When the 880 nm LED source was 103 introduced in the Aethalometers AE16, AE21, AE22, AE31 and AE42, the mass attenuation cross 104 section at this wavelength was determined by comparison with older-type Aethalometers to be 16.6 105 m²g⁻¹ (Hansen, 2005).

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107 The path length of a photon and hence the probability for the photon to be absorbed by a particle 108 increases in the filter matrix due to the scattering of light by the filter fibers. This can be empirically 109 taken into account using a single parameter, describing the enhancement of absorption as a 110 multiplication factor C (Weingartner et al., 2003): $\sigma = C \sigma_{air}$. The same mass of the sample absorbs C-111 times more when the particles are embedded in the filter matrix than when they are freely suspended in the air. The separation of the C and σ_{air} parameters, using the value of C = 2.14 112 (determined in Weingartner et al., 2003) for legacy type Aethalometers, such as AE31, determines 113 the mass absorption cross-section of freshly emitted BC σ_{air} = 7.77 m²/g. We use this parameter in the 114 115 AE33 in addition to the parameter C, determined for the new TFE-coated glass fiber filter tape (see section "3.3 Influence of the filter material"). We measured ambient BC concentrations using 116 117 collocated Aethalometers AE31 and AE33, then determined the parameter C for the AE33 from these 118 ambient data.

120 Optical measurements in the infra-red part of the spectrum should be used to convert the optical 121 measurement into the mass concentration, as the contribution of sample components other than 122 black carbon is negligible at these wavelengths (Sandradewi et al., 2008a; Sandradewi et al., 2008b; 123 Fialho et al., 2005; Yang et al., 2009; and references therein). The relationship between the mass 124 concentration of BC and the optical absorption can be determined by comparing the filter 125 photometer measurements with those obtained by thermal-optical analysis (Sciare et al., 2011). 126 However, as the determination of elemental carbon (EC) depends on the thermal-optical analysis 127 method, sometimes with large differences (Bae et al., 2009), the determination of the mass 128 absorption cross-section also depends on the thermal-optical method employed. Additionally, the season and the sample composition (Bae et al., 2009; Chiappini et al., 2014) may influence the 129 130 determination of EC; and the mass absorption cross-section may depend on the aerosol mixing state 131 and size (Bond and Bergstrom, 2006). Concurrent determination of EC in filter samples with an 132 thermal-optical method, and the Aethalometer measurement of optical absorption allow the sitespecific determination of the mass absorption cross-section, which is specific to the thermal-optical 133 method employed – this procedure is often employed in source apportionment campaigns, where 134 135 mass closure is attempted (Sciare et al., 2011).

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137 Determination of the face velocity ratio factor (FVRF)

The loading effect compensation algorithm is sensitive to correct measurement of the sample flow through the spots. The flow through the spot2 is especially susceptible to errors since it is calculated as a difference between total flow and flow through the spot1. *FVRF* is used to compensate for flow measurement uncertainty (Article, Equation 11):

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$$FVRF = \left(\frac{ATN_{spot2}}{ATN_{spot1}}\right)_{ATN=0} \frac{F_1}{F_2}$$

The calculation of *FVRF* is based on the fact that at small filter loadings the attenuation is proportional to the flow through the spot. Because of the transients on the fresh spot, the first *ATN* measurements (where $ATN_1 < ATN_{f_1}$) are omitted from the analysis. Data with ATN_1 (attenuation for channel1, spot1) between the lower limit ATN_{f_1} and upper limit ATN_{f_2} are used for determination of *FVRF* (Figure S6). The actual parameter which tells us the correct flow ratio is the intercept of the linear fit of ATN_{spot2}/ATN_{spot1} versus ATN_{spot1} (Figure S6). The average *FVRF* for channels 2 to 6 is used for further calculations.



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FVRF values differ slightly between the instruments and spots. Here is an example of mean values and standard deviation of *FVRF* for 5 different instruments: 1.001 ± 0.025 , 1.059 ± 0.011 , $1.063 \pm$ 0.001, 1.017 ± 0.006 and 0.979 ± 0.02 . If the flow ratio is not compensated, a wrong value of parameter *k* is obtained. The influence of 2 % change of flow ratio on the parameter *k* is presented in Figure S7.





160 Figure S7. Illustration of the sensitivity of a dual spot loading effect compensation algorithm to

161 a 2% change in the face velocity ratio factor (FVRF).

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163 Weighing method for loading effect compensation parameter *k*

Determination of the parameter *k* at low filter loadings is very susceptible to small measurement errors. When more and more material is collected on the spot, the filter loading effect is easier to measure using the dual spot approach. To reduce the uncertainty of the parameter *k* at low attenuations, a weighting method is applied (main body of the article, Equation 12). Both weighted and un-weighted values of parameter *k* are presented on Figure S8.

- 169 For $ATN_1 < ATN_{f2}$ a value of k_{old} , which is obtained from the previous filter spot, is reported:
- 170 $k_{weighted}(ATN_1 < ATN_{f2}) = k_{old}.$

For $ATN_1 > ATN_{f_2}$ a weighted value of the parameter k is used, which takes into account the k_{old} and the un-weighted value of parameter k. The default value is $ATN_{f_2}=30$. Progressively, as the attenuation increases, less and less of k_{old} is incorporated into $k_{weighted}$:

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$$k_{weighted} = \frac{(ATN_{TA} - ATN_1)k_{old} + (ATN_1 - ATN_{f_2})k}{(ATN_{TA} - ATN_{f_2})}.$$

175 At the time of full spot loading, the weighted value of k becomes equal to the instantaneous (un-

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$$k_{weighted}(ATN_1 = ATN_{TA}) = k$$

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Figure S8. Example of the instantaneous and the weighted value of the parameter k during spot loading. For $ATN_1 < ATN_{f2}$ the k value from the previous spot is used. For attenuations between $ATN_{f2}=30$ and $ATN_{TA}=120$, a weighting method is applied.



185 Figure S9. Time-series of the ACTRIS inter-comparison at TROPOS, showing AE33 and MAAP

186 measurements of BC.

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