Aerosol light absorption from attenuation measurements of PTFE-membrane filter samples: implications for particulate matter monitoring networks

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Abstract. Mass absorption cross-section (MAC) measurements of atmospherically-relevant aerosols are required to quantify their effect on Earth's radiative budget. Estimating aerosol light absorption from transmittance and/or reflectance measurements through filter deposits is an attractive option because of ease of deployment in field settings and low cost. These measurements suffer from artifacts that depend on a given filter measurement system and type of aerosol. Empirical correction algorithms are available for commercial instruments equipped with optically-thick fiber filters, but optically-thin filter media have not been characterized in detail. Here, we present empirical relationships between particle light absorption–measured using multi-wavelength integrated photoacoustic spectrometers and nephelometers–and attenuation measurements for polytetrafluoroethylene (PTFE) membrane filter samples of carbonaceous aerosols generated from combustion of diverse biomass fuels and kerosene (surrogate for fossil-fuel combustion). We establish a simple, wavelength-independent formulation for calculating aerosol MAC and absorption coefficients from filter attenuation measurements. We find the ratio between in-situ absorption and bulk attenuation to be inversely proportional to aerosol single scattering albedo. As a case study, we apply our formulations on 2010 attenuation datasets of the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network to quantify the overestimation in their filter-based absorption coefficients.

1 Introduction

Aerosol light absorption affects the radiative balance of the Earth's atmosphere through direct and indirect mechanisms (Bond et al., 2013; Kanakidou et al., 2005; Ramanathan et al., 2001). The light absorption metric relevant to climate modelers–mass absorption cross-section (MAC)–depends on the size, shape and composition of the aerosols (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006; Moosmüller et al., 2009). This property has a complex dependency on the emission source, especially for carbonaceous aerosols (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006; Chakrabarty et al., 2010).

A first-principle method of measuring contact-free aerosol light absorption is using photoacoustic spectroscopy, which employs lasers at selected wavelengths to heat the aerosols, thereby producing a detectable pressure signal (Arnott et al., 1999). Absorption can also be estimated as the difference between *in-situ* measurements of extinction and scattering

(Schnaiter et al., 2005; Sheridan et al., 2005). Alternatively, a commonly adopted technique for estimating light absorption uses measurements of transmittance and/or reflectance for aerosol particles collected on a filter substrate. Instruments developed based on this technique, including the aethalometer (Hansen et al., 1984) and the Particle Soot Absorption Photometer or PSAP (Virkkula et al., 2005), facilitate semi-continuous sampling of particles and produce time-averaged bulk absorption measurements. Particles may also be collected on quartz fiber or Teflon filters and analyzed for their absorption using standalone spectrophotometers (Pandey et al., 2016; White et al., 2016; Zhong and Jang, 2011).

Filter-based measurements are attractive because of their ease of deployment in field settings and low cost, but they suffer from several artifacts. Particles embedded in a multiple-scattering medium experience a larger optical path length than particles in their native suspended state, leading to the appearance of enhanced light absorption (Bond et al., 1999; Clarke, 1982; Gorbunov et al., 2002). This is referred to the as the multiple scattering artifact, and it depends on the choice of filter medium. A higher loading of absorbing aerosols can diminish the effect of multiple scattering, inducing an aerosol dependent loading artifact (Arnott et al., 2005; Weingartner et al., 2003). Highly scattering aerosols could enhance multiple scattering and lead to increased backscatter, which leads to an overestimation of absorption (Lack et al., 2008; Weingartner et al., 2003). These artifacts have been evaluated for several commonly used filter-based instruments, such as those aforementioned, by comparing their measurements with contact-free aerosol light absorption measurements or using reference materials with known optical properties. Typically, correction algorithms for these artifacts are formulated as functions of some combination of filter and aerosol properties (Arnott et al., 2005; Collaud Coen et al., 2010; Virkkula, 2010; Weingartner et al., 2003) and are specific to a given measurement system.

In various field settings, aerosol samples are collected on polytetrafluoroethylene (PTFE) membrane filters (commonly known as Teflon filters) for inferring ambient or near-source particulate mass concentrations using gravimetric analysis (Koistinen et al., 1999). Major aerosol monitoring networks, such as the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network (Chow et al., 2010; Solomon et al., 2014), the Chemical Speciation Network (CSN) (Solomon et al., 2014) and the Surface PARTiculate mAtter Network (SPARTAN) (Snider et al., 2015), collect particle samples on Teflon filters for gravimetric and elemental measurements. PTFE filters are chemically inert and unlike quartz fiber filters, present a very low surface area for organic vapor adsorption (Kirchstetter et al., 2001; Vecchi et al., 2014). Correction schemes developed for instruments that use fiber filters (like the PSAP and aethalometer) cannot be applied to infer aerosol light absorption properties using measurements of transmittance and/or reflectance on PTFE filters. A previous study on the artifacts associated with this estimation used a reference material and provided a constant multiple scattering correction factor for optical loadings smaller than a certain threshold (Zhong and Jang, 2011). Another recent study (White et al., 2016) proposed a theory-based model to calibrate attenuation measurements for Teflon filter samples and applied this new model to a historical dataset from IMPROVE network. They found that the reevaluated absorption values for the PTFE samples were well-correlated with thermo-optical elemental carbon (EC) measurements for co-located quartz fiber filters.

In this work, we generated carbonaceous aerosols with varying physicochemical properties from the combustion of biomass fuels and kerosene. Combustion conditions were varied to yield a range of intrinsic aerosol optical properties. Kerosene

combustion was used as a surrogate for fossil fuel burning, which is linked with soot or EC emissions (Andreae and Gelencsér, 2006; Bond et al., 2013). The combustion of wildland- and fuel-biomass is implicated in emissions of EC as well as light absorbing organic carbon (LAOC) (Andreae and Gelencsér, 2006; Chakrabarty et al., 2010; Chen and Bond, 2010). EC is known to absorb light throughout the visible and UV wavelengths, while LAOC absorbs preferentially in the near-UV

5 and UV regions (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006; Kirchstetter et al., 2004; Sun et al., 2007). Therefore, we measured *in-situ* and contact-free aerosol light absorption and scattering coefficients using integrated photoacoustic-nephelometer (IPN) spectrometers operated at three wavelengths - 375, 405 and 532 nm. Co-located with these measurements was a sampling system to collect particles onto Teflon membrane filters. Subsequent measurements of light attenuation, using ultraviolet-visible (UV-vis) spectrophotometer, were performed on the filter samples. Observed

10 empirical relationships between particle light absorption and filter attenuation were established in conjunction with predictions from a one-dimensional (1-D) two-stream radiation transfer model.

2 Methods

2.1 Experiments

Diverse biomass fuels including wood and needles from pine, fir and sage trees, grass, peat and cattle dung were burned in a

15 21 m^3 stainless steel combustion chamber located at Washington University (Sumlin et al. (2017); Sumlin et al. (2018)). Flaming, smoldering and mixed combustion phases were employed (see Supplement) to generate a range of intrinsic aerosol properties: single scattering albedo (SSA) values at 375, 405 and 532 nm ranged 0.25-0.99 and Absorption Ångström Exponents (AÅE) for 375-532 nm ranged 1.2-6.8. A kerosene lamp was used to generate soot particles, with an SSA of 0.3 and AÅE within 0.70-1.1. A schematic of the experimental setup is shown in Figure 1. Approximately 10-50 g of a given

20 type of woody biomass/grass/dung was placed in a stainless-steel pan and ignited using a flame. It was either allowed to continue flaming or brought to a smoldering phase by starving the flame with a lid. In the same type of pan, 5-15 g of peat was smoldered by using a ring heater to raise its temperature to 200 □C. In one set of experiments, smoke from the chamber was directly sampled, while in another set, a hood placed over the pan was used for sampling the aerosols. The chamber exhaust was closed during the burns. The outlet from the hood or chamber was passed through a diffusion dryer and a semi-

25 volatile organic compound (SVOC) denuder into a mixing volume, from which aerosols were continuously sampled by the four IPNs.

During each burn, optical (absorption and scattering) signals were monitored using IPNs until a steady state was reached. During the steady state, particle samples were collected on 47 mm PTFE membrane (Pall) filters. The filter sampling flow rate was set to 5 liters per minute and the sampling durations were between 2 and 20 minutes. For each filter sample, an

30 absorption optical depth ($\tau_{a,s}$) of the deposited aerosols was calculated from the absorption coefficients measured using the IPNs:

$$\tau_{a,s} = \frac{b_{abs,av} \times Q \times t_s}{10^9 A_s} \tag{1}$$

where $b_{abs,av}$ is the average absorption coefficient (in Mm^{-1}) during the sampling duration t_s (in min), Q is the flow rate (in liters per minute or lpm) through the filter and A_s is the filter sample area (in m^2). Optical depth $\tau_{a,s}$ for the samples in this study ranged between 0.01 and 0.68. The uncertainty in these estimates was predominantly from the standard deviation in

5 $b_{abs,av}$ over the averaging interval, and was within 10% for all samples.

Transmittance (T) and reflectance (R) for the filter samples were measured using a Perkin-Elmer LAMBDA 35 UV-vis spectrophotometer (described in Zhong and Jang (2011)). Attenuation (ATN) through the filter samples was calculated using (Bond et al., 1999; Campbell et al., 1995):

$$ATN = ln(\frac{1-R}{T}) \tag{2}$$

10 When this equation is applied to blank filters, it results in ATN values between 0.01-0.03. A wavelength dependent "blank attenuation" was subtracted from the sample attenuation values. Replicate transmission and reflection measurements were used to estimate measurement error; these yielded an uncertainty of 5% in the calculated attenuation.

A correction factor (C) that captures the net effect of multiple scattering and aerosol loading can be defined as:

$$\tau_{a,s} = C \times ATN \implies C = \frac{\tau_{a,s}}{ATN} \tag{3}$$

15 **2.2 Two-stream radiative transfer model**

A 1-D two-stream radiative transfer framework for multiple scattering in absorbing media was developed in Bohren (1987) and subsequently discussed in relation to aerosol-filter systems in several studies (Arnott et al., 2005; Clarke, 1982; Gorbunov et al., 2002; Petzold and Schönlinner, 2004). Solving a radiation balance for an aerosol-laden filter medium yields the following expressions for transmittance (T_l) and reflectance (R_l), respectively:

20 $$T_l = \frac{2}{[2K - \omega_l(1-g_l)\,sinh(K\tau_{e,l}) + 2\,K\,cosh(K\tau_{e,l})]} \tag{4A}$$

$$R_l = \frac{\omega_l(1-g_l)\,sinh(K\tau_{e,l})}{[2K - \omega_l(1-g_l)\,sinh(K\tau_{e,l}) + 2\,K\,cosh(K\tau_{e,l})]} \tag{4B}$$

Here, ω_l, g_l and $\tau_{e,l}$ denote the SSA, asymmetry parameter and extinction optical depth, respectively, of the composite layer. The parameter K is defined as:

$$K = \sqrt{(1 - \omega_l)(1 - g_l\omega_l)} \tag{5}$$

25 Arnott et al. (2005) used the above model to derive the form for an approximate correction factor for the aethalometer. The aethalometer uses optically-thick quartz fiber filters, which are strongly multiple scattering, transmitting only ~10% of light in the visible wavelengths. A mathematical consequence of strong multiple scattering is that the term $K\tau_{e,l}$ is much greater

than unity and Eq.s (4A)and (4B) can be replaced by simplified approximations. In contrast, the Teflon filters used in this study are optically thin and constitute a weak multiple scattering medium: they transmit 70-80% of incident visible light. Therefore, the full equations for T_l and R_l were solved for the filter-particle system, using a range of values of $\tau_{a,s}$ and SSA consistent with experimental observations. Two other required inputs could not be measured: the penetration of aerosols into

5   the filter was assumed to be 10%, and the asymmetry parameter of the aerosols was fixed at 0.6, based on the typical values reported for biomass burning emissions (Martins et al., 1998; Reid et al., 2005). Transmittance and reflectance through a two layer system – the aerosol laden layer with properties T_l and R_l and a pristine filter layer with properties T_f and R_f – were calculated (Gorbunov et al., 2002):

$$T = \frac{T_l T_f}{1 - R_l R_f} \tag{6A}$$

10    $$R = R_l + \frac{T_l^2 R_f}{1 - R_l R_f} \tag{6B}$$

Using the above results, ATN was calculated (per Eq. (2)), and the results were used to examine the relationship between the properties of the aerosol deposits and the attenuation of light through the two-layer composite system. Further details of the calculations in this section are provided in the Supplement.

3 Results and discussion

15   Modeled and experimental values of light attenuation through filter samples are shown in Fig. 2. Certain combinations of SSA and $\tau_{a,s}$ (shaded region in the figure) were never observed in our experiments (black dots): high SSA aerosols are associated with lower absorption per unit mass, therefore very high mass loadings would be required to yield the upper range of the $\tau_{a,s}$ in this study. For SSA<0.9, the modeled attenuation values show little spread with changing SSA. Like the model predictions, experimental data show a non-linear nature relationship between attenuation and aerosol absorbance. However,

20   attenuation values calculated from measurements are slightly lower than model predictions. This may be due to differences between assumed parameters in our model and their deviation from real-world values.

The well-constrained relationship between aerosol and filter measurements in Fig. 2 suggests that $\tau_{a,s}$ of deposited particles could be directly estimated by measuring light attenuation. The best fit relationship ($R^2 = 0.87$) between both parameters is given by:

25    $$\tau_{a,s} = 0.48 \, (ATN)^{1.32} \tag{7}$$

In Fig. 3, we combined all experimental data corresponding to the three wavelengths since our measurements showed no clear stratification with varying wavelength. Also shown in the figure are estimated $\tau_{a,s}$ using a constant correction factor C of 0.67 proposed by Zhong and Jang (2011) (black perforated line); this correction factor clearly overestimates $\tau_{a,s}$ for most ATN values investigated in this study. We find our data to be better represented by an approximate $C = 0.46$ based on a

linear least-squares fit ($R^2 = 0.79$). However, any constant C value does not capture the non-linearity of the interaction between aerosol properties and the multiple-scattering within the filter medium. It should be noted that C in Eq. (3) represents the net effects of all filter artefacts. There are measurement errors associated with both ATN and $\tau_{a,s}$, and therefore, C contains propagation of uncertainties from both parameters. There was no correlation between C and ATN (see

5   Fig. S3). We observed an inverse relationship between C and SSA (Fig. 4), consistent with results the from the two-stream radiative transfer model. For a given value of $\tau_{a,s}$, measured ATN will always be higher for aerosols with higher SSA values. Consequently, we should expect C to decrease with increasing SSA; this decreasing relationship for our experimental data is given by:

$$C = -0.76 * SSA + 1.02 \tag{8}$$

10   Values of C and SSA for individual samples (shown in supplemental Fig. S3B) were aggregated into five SSA bins to demonstrate the inapplicability of an empirical correction factor formulation to low SSA data points in this study. The large spread in C values for low SSA is likely due to noise amplification from dividing two small ($\tau_{a,s}$ and ATN < 0.2) numbers. For SSA>0.6, the correction factor decreases linearly.

4 Implications for aerosol monitoring networks

15   Teflon filters are routinely used for gravimetric and elemental analysis across monitoring networks (Chow et al., 2010; Snider et al., 2015; Solomon et al., 2014), as well as field and laboratory source characterization studies. The $\tau_{a,s}$ versus ATN relation from Eq. (7) was applied to IMPROVE network's attenuation dataset from the year 2010, representing samples collected at 223 sites (details in Supplement). Their measurements were carried out at 633 nm wavelength using a Hybrid Integrating Plate and Sphere (HIPS) method (Bond et al., 1999). A detailed description of the measurement technique can be

20   found in White et al. (2016). At each of these sites, co-located quartz filters were used to measure EC mass concentrations using thermo-optical analysis (Chow et al., 2007). EC is considered to be a surrogate for light absorbing aerosol species (Chow et al., 2010) and is therefore expected to correlate with estimates of light absorption. The HIPS-measured ATN data, after being corrected using our formulation showed a slightly improved correlation with corresponding EC measurements than the uncorrected measurements (Fig. S4). The ratio of uncorrected to corrected HIPS filter absorption coefficients (Fig.

25   5) is inversely correlated with EC mass concentrations. This is consistent with model predictions (Fig. 1) and experimental data (Fig. S3) that show that the relative contribution of the filter medium to ATN is high when $\tau_{a,s}$ is small. Overall, Fig. 5 shows that for lightly loaded filters (EC concentration < 0.1 μgm^{-3}), uncorrected filter ATN measurements could lead to a 4- to 16-fold overestimation in the inferred absorption coefficients.

5 Conclusions

30   We evaluated the relationship between *in-situ* aerosol light absorption and attenuation of aerosol deposits on Teflon filters for combustion aerosols (encompassing $0.25 \leq SSA \leq 0.99$), at 375, 405 and 532 nm wavelengths. An empirical non-linear relationship was found between the absorption optical depth of sampled aerosols and attenuation through filter samples; the

nature of this function was consistent with predictions from a two-stream radiative transfer model of the filter-aerosol system. Following Eq. (7), we propose the estimation of Aerosol MAC (m^2g^{-1}) values from filter ATN measurements using:

$$MAC = [0.48 \, (ATN)^{1.32}] \frac{A_s}{m} \qquad (9)$$

where A_s is the filter sample area (in m^2) and m is the mass on deposited particles (in g). Additionally, aerosol absorption coefficients (b_{abs}; in Mm^{-1}) can also be calculated using:

$$b_{abs} = [0.48 \, (ATN)^{1.32}] \frac{10^9 A_s}{Q \times t_s} \qquad (10)$$

The quantities Q and t_s are as used in Eq. (1). Caution must be taken, as suggested by the two-stream model results, on the limits of applicability of the empirical relationships (equations 7-10)–significant errors could result from application of the relationships if the aerosol SSA>0.9 and ATN values are beyond the range of this work.

Teflon filters are routinely used for gravimetric and elemental analysis, across aerosol monitoring networks, as well as field and laboratory source characterization studies. Therefore, we applied this Eq. (7) to attenuation data from all IMPROVE sites for the year 2010 and found that there was a slightly improved correlation with independent measurements of EC mass concentration. For low aerosol concentrations, the measured attenuation coefficients may be 4-16 times larger than the aerosol absorption coefficient.

Supplement

Includes a schematic of the experimental setup and table of experiments (Text S1, Figure S1, Table S1), detailed equations for the two-stream model (Text S2, figures S2 and S3), correction factor plots (Text S3, figures S4 and S5), and addition information on the implications (Text S4 and Figure S6).

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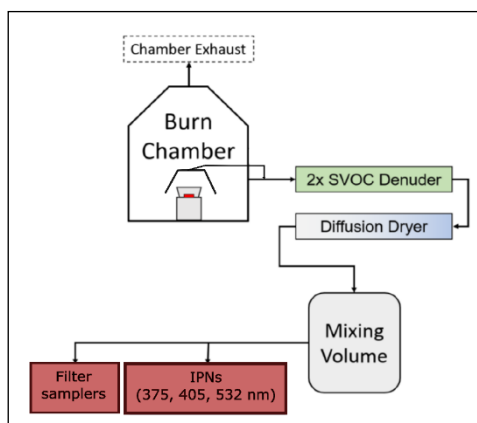
Figure 1: Schematic representation of the experimental setup. Inlet to the semi-volatile organic compound denuder was taken from either the chamber sampling port or the hood.
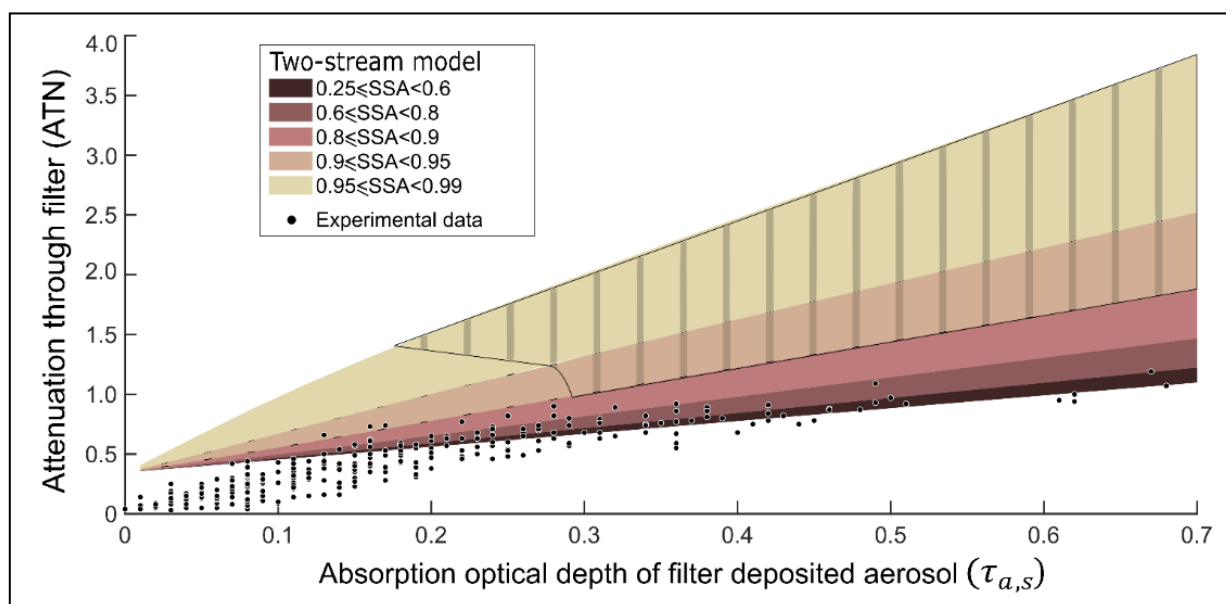
Figure 2: Attenuation through a modeled two-layer aerosol laden filter as a function of the absorption optical depth and single scattering albedo of the aerosols. The shaded portion represents aerosol properties not relevant to the measurements in this study. Experimental data (black circles) are overlaid.
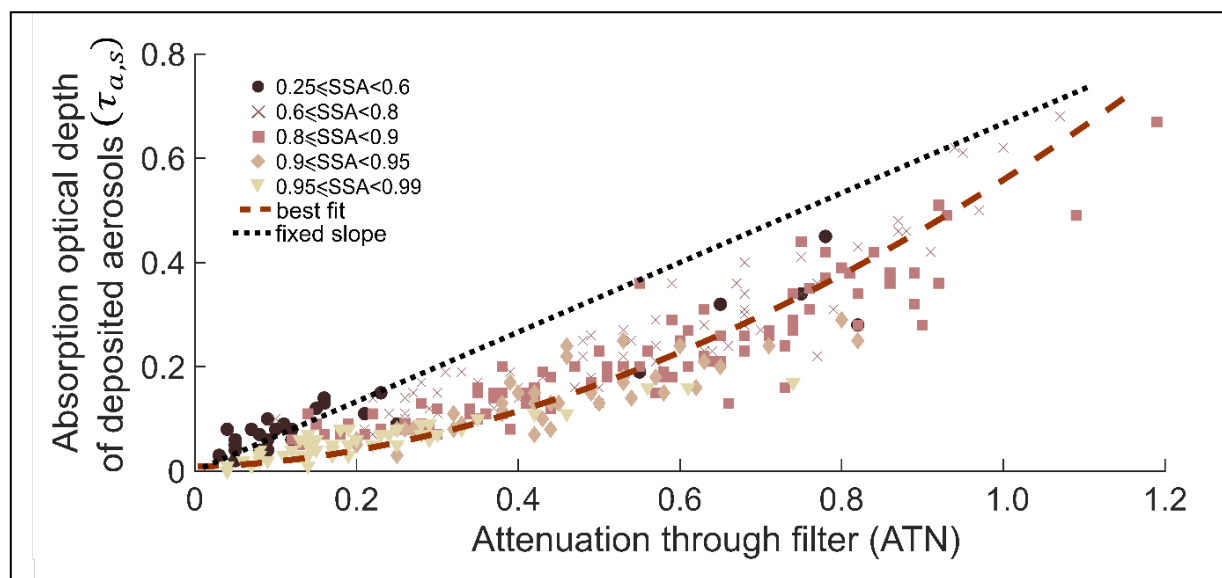
Figure 3: Relationship between absorption optical depth and attenuation through carbonaceous aerosol filter sample measured at 375, 405 and 532 nm. The best fit curve is given by Eq. (7). The black perforated line has a fixed slope of 0.67 per Zhong and Jang, 2011.
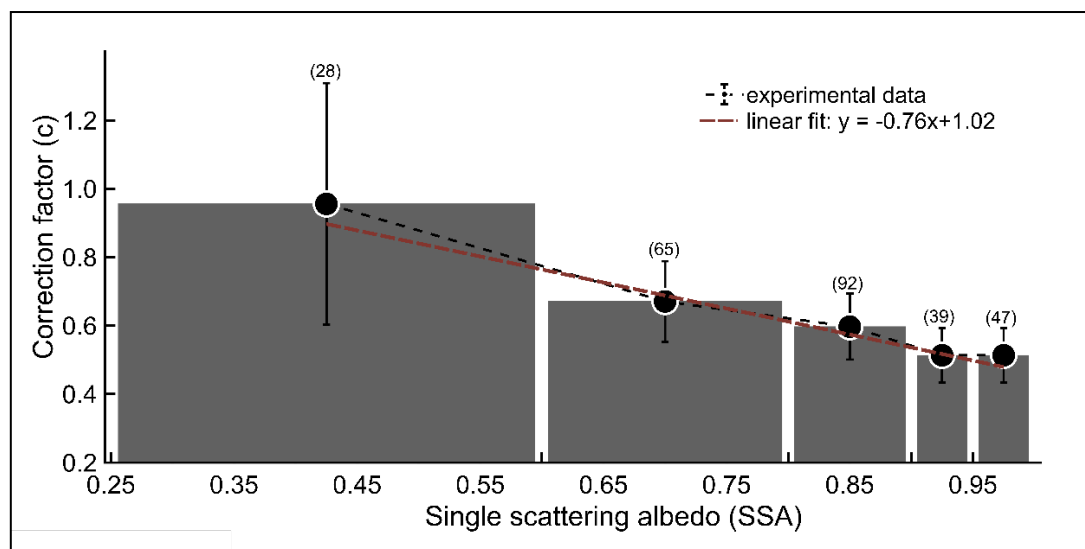
Figure 4: Correction factor C for filter artefacts as a function of single scattering albedo of the deposited aerosols. Error bars show one standard deviation around the mean. The numbers in parentheses denote the number of data points in each bin.
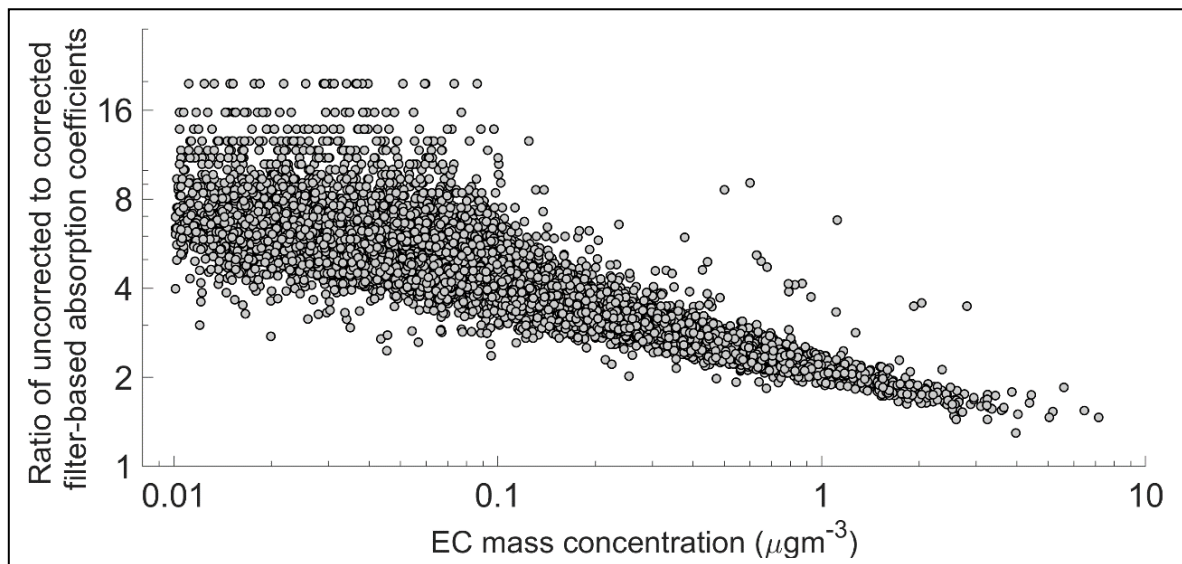
Figure 5: Ratio of uncorrected filter absorption coefficients at 633 nm, measured from IMPROVE samples to corrected absorption coefficients obtained using Eq. (7), as a function of EC mass concentration. Absorption coefficients are from attenuation measurements on Teflon filters and EC concentrations are from thermo-optical measurements on quartz filters; IMPROVE network collects samples every 3 days at 223 sites.