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An accurate filter loading correction is essential for assessing personal exposure to black carbon using an Aethalometer

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Abstract

The AE51 micro-Aethalometer (microAeth[®]) is a popular and useful tool for assessing personal exposure to particulate black carbon (BC). However, few users of the AE51 are aware that its measurements are biased low (by up to 70%) due to the accumulation of BC on the filter substrate over time; previous studies of personal black carbon exposure are likely to have suffered from this bias. Although methods to correct for bias in micro-Aethalometer measurements of particulate black carbon have been proposed, these methods have not been verified in the context of personal exposure assessment. Here, five Aethalometer[®] loading correction equations based on published methods were evaluated. Laboratory generated aerosols of varying black carbon content (ammonium sulfate, Aquadag[®] and NIST diesel particulate matter) were used to assess the performance of these methods. Filters from a personal exposure assessment study were also analyzed to determine how the correction methods performed for real-world samples. Standard corrections equations produced correction factors with root mean square errors of 0.10 to 0.13 and mean bias within ± 0.10 . An optimized correction equation is also presented, along with sampling recommendations for minimizing bias when assessing personal exposure to BC using the AE51 micro-Aethalometer.

1. Introduction

The association between exposure to fine particulate matter (PM) and adverse health outcomes has been demonstrated in a number of studies (1). The evidence is more limited, however, for linking specific components of fine PM to adverse health (1). One PM component of particular interest is black carbon (BC), because BC is marker of combustion-derived PM and because exposure to combustion-derived PM has established associations with adverse human health (2, 3).

Human exposure to BC is typically quantified using optical techniques (e.g. 4, 5). This work investigates the Aethalometer[®], one of the most common techniques used to assess personal BC exposure. An Aethalometer measures the attenuation in light intensity (I) through an air

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sampling filter with respect to a reference beam (I_0), from which an equivalent BC concentration can be derived (6). The microAeth[®] Model AE51 (AethLabs, San Francisco, CA, USA) is the only commercially available Aethalometer small enough for on-person measurements, produces results consistent with a full-size Aethalometer (7, 8), and hence is the de-facto instrument for assessing personal BC exposure. The AE51, the focus of this work, operates at 880nm, uses Teflon coated glass fiber (Pallflex[®] Fiberfilm[™] T60A20, Pall Life Sciences, MI, USA) filters, samples at 50 to 250 ml/min of flow with a logging interval as fast as 1 Hz, weighs ~280 g and runs off battery power.

The percent attenuation (ATN) derived by an Aethalometer ($100 \times \ln(I_0/I)$) is dependent upon both scattering and absorption of light by a PM sample that accumulates onto the filter matrix. By measuring the change in ATN (ΔATN) over time, the AE51 is able to estimate the accumulation of BC onto an air sampling filter, and hence, ambient concentration of BC in air. Relating the change in ATN to an appropriate BC concentration becomes complicated, however, as the filter becomes increasingly loaded with PM; both scattering and absorption phenomena must be accounted for to derive the instantaneous absorption coefficient from which a BC concentration can be calculated (9–12). Light scattered by the filter and embedded particles will alter the measured ATN by increasing the optical path and scattering light away from the detector. The accumulation of absorbing particles on the filter decreases the effective optical path resulting in a loading dependent relationship between the measured ATN and BC mass. High filter loading also make BC concentration derivation unreliable, AE51 reports the filter is overloaded when ΔATN reaches 125, however the maximum reliable ΔATN may be nearer 75 (6, 13).

Personal exposure studies often report no correction for filter loading of the AE51 (e.g. 14, 15–26). Several correction equations have been proposed to compensate for measurement artifacts associated with the Aethalometer (27). However, only the basic empirical corrections, which do not explicitly account for variations in aerosol properties such as scattering, tend to be applied in personal exposure studies (e.g. 28, 29–32). Failure to account for the loading of absorbing particles onto the filter will bias the measurements low i.e., the derived BC concentration will be increasingly underestimated with increased filter loading (measured BC is less than the actual BC concentration). Failure to account for changes in particle light scattering will also lead to measurement error. Furthermore, BC particles may be present in varying fractions as internally (BC combined with other material in a single particle) and externally mixed (BC only) particles, even in urban environments (e.g. 33). The magnitude and direction of bias from the accumulation of non-BC particles and mixing state onto the AE51 filter has not been investigated.

Motivated by the above discussion, this work aimed to quantify the appropriateness of AE51 correction factors by assessing their ability to reduce bias due to the cumulative loading of absorbing PM during a measurement. Based on the assessment of existing correction factors, optimized coefficients and a validated loading correction equation were developed to reduce bias associated with AE51 use in personal exposure studies. The correction equations described here could also be applied to previously collected BC exposure data.

2. Methods

The study was designed with three primary aims. The first aim was to determine the measurement error in BC concentration as a function of BC loading onto an AE51 filter substrate. The second aim was to evaluate the performance of loading correction equations commonly applied to AE51 data. The third aim was to investigate if newly-derived fitting coefficients (based on actual personal BC exposure data) or a newly derived correction equation would improve personal BC exposure measurement using the AE51.

The BC loading effect was assessed by comparing the AE51's response with a loaded filter to that with a fresh, unloaded filter substrate (as a reference) in a controlled laboratory setting. However, because a constant reference concentration is difficult to maintain (even for a laboratory chamber experiment), an online photoacoustic extinctionsmeter (PAX, Droplet Measurement Technologies, Boulder, CO, USA) (34) was used to account for minute-to-minute variations in BC concentration during an experiment. According to Equation 1, R_{meas} is the ratio of an AE51 BC measurement using a loaded filter to that of an unloaded filter (the reference measurement), corrected for temporal variations in BC concentrations by the PAX. Thus, R_{meas} is the fractional error in BC concentration reported by the AE51 due to the accumulation of BC onto the filter substrate over time:

$$R_{meas} = \frac{BC_{AE51}}{BC_{PAX}} \times \frac{BC_{PAX,Ref}}{BC_{AE51,Ref}}; \quad (1)$$

On the right-hand side of Equation 1, BC_{AE51} and BC_{PAX} are the BC concentrations measured concurrently by the AE51 and PAX, $BC_{AE51,Ref}$ is the BC concentration measured using an unloaded filter, and $BC_{PAX,Ref}$ is the PAX BC concentration measured concurrently with $BC_{AE51,Ref}$. Thus, R_{meas} equals one when the AE51's filter is unloaded (or when the BC loading effect produces negligible error in the measurement). The PAX method does not require a filter (and therefore does not suffer from loading artifacts) and also includes a scattering measurement so that aerosol extinction (scattering and absorption) at a wavelength of 870 nm can be measured. The percent error in measured BC concentration due to loading is then given by equation 2:

$$percent\ error = (1 - R_{meas}) \times 100; \quad (2)$$

The performance of five loading correction equations was compared by calculating difference in the measured (R_{meas}) and predicted (R_{eqn}) change in BC concentration, according to equation 3:

$$error = (R_{meas} - R_{eqn}); \quad (3)$$

The overall performance of each loading equation was assessed via the root mean square error (RMSE) and the mean bias (MB), according to equations 4 and 5, where n is the

number of measurements. Corrections with lower RMSE and MB closer to zero were judged to perform better.

$$RMSE ; = \sqrt{\frac{\sum (error^2)}{n}} \quad (4)$$

$$MB ; = ; \frac{\sum error}{n} \quad (5)$$

This work characterized the response of the AE51 under a range of realistic filter loading conditions and assessed the validity of five correction algorithms that account for the cumulative effect of absorbing particles sampled onto the AE51 filter. Two types of aerosol chamber experiments were conducted to assess the loading effect. In the first type, particles of known composition were sampled onto new filter substrates. The performance of the AE51 (as a function of filter loading) was then evaluated against a reference measurement. In the second type, AE51 filters that were used during the Fort Collins Commuter Study (35) (i.e., filters pre-loaded with varying amounts of PM) were loaded with additional (known) quantities of BC. Thus, the effectiveness of loading correction methods could be assessed for different particle types and for actual personal measurements taken across a range of exposure levels.

2.1 Aerosol Generation

Aerosols were generated continuously inside a 0.75m³ chamber using a single-jet Collison nebulizer (model BGI CN24, Mesa Labs Inc.). The chamber was purged with dry air (< 10% RH) to evaporate water from the nebulized droplets. A stable concentration of particles was maintained by controlling the purge flow rate through the chamber (~60 L/min). An over-pressure of ~ 0.1" H₂O was maintained in the chamber to prevent extraneous particles being sampled. The aerosol was mixed into the chamber using a fan located in the corner.

2.2 Sampling Setup

Aerosol was sampled from the center of the chamber through a 1 m stainless steel tube (ID = 0.89 cm) and split between the AE51s, a PAX and a scanning mobility particle sizer (SMPS) (model 3081 classifier, 3787 particle counter, TSI Inc., Mn, USA). Chamber temperature (°C) and relative humidity (%) were continuously measured using a compact data logger positioned near the center (model MSR145/FT/FH/020, MSR Electronics GmbH., CH). The chamber sampling set-up is illustrated in Figure 1.

2.3 Photoacoustic BC Measurement (PAX)

Reference BC measurements were determined using a photoacoustic extinctions (PAX). The PAX directs a ($\lambda = 870$ nm) laser beam across its sample chamber modulated at the resonant frequency of the chamber. The modulated beam heats absorbing material which in turn heats the surrounding air to produce a sound pressure wave. The properties of this

pressure wave (resonator quality factor Q , and resonance frequency f) are measured by a microphone. The volumetric mass concentration of BC is then derived from the measured absorption coefficient (B_{abs}) by assuming a particle absorption cross-section (34). An advantage of the photoacoustic technique for BC measurement over filter-based methods is that there is no filter-loading artifact; thus, if the unloaded-Aethalometer to PAX ratio is known for a given aerosol type, the PAX can be used as a reference to quantify the filter loading effect within the Aethalometer. The PAX simultaneously measures the aerosol scattering coefficient (B_{scat}) ($\lambda = 870$ nm) using reciprocal nephelometry; thus, the fraction of aerosol extinction due to scattering (or single scattering albedo, ω) can also be calculated.

2.4 Preliminary correction factor validation

Four types of preliminary experiments were performed. The first set of experiments utilized ammonium sulfate, a purely scattering aerosol, to quantify the effects on non-BC particles on the AE51. For these tests, a 1 g/L ammonium sulfate (ACS grade, Fisher Scientific, MA, USA,) solution was nebulized and sampled for ~20 hours (target PM concentration of 10 $\mu\text{g}/\text{m}^3$). Second, a solution of Aquadag® (Acheson Inc., MI, USA), was nebulized into the chamber to serve as a proxy for BC loading onto filters (36, 37). Aquadag comprises of fragments of thin plates of crystalline graphite (38). Aquadag experiments (target PM concentration 10 $\mu\text{g}/\text{m}^3$) were performed to investigate the BC loading effect up to ATN values of 125 (nominally the upper range of the AE51). Third, NIST Standard Diesel Particulate Matter (1650b, NIST, Gaithersburg, MD, USA) was generated (target BC concentration 10 $\mu\text{g}/\text{m}^3$) in the chamber to compare the Aquadag loading effects to a more complex aerosol. Finally, mixtures of Aquadag and ammonium sulfate were nebulized simultaneously to investigate the single scattering albedo dependence (target particle concentration 20 $\mu\text{g}/\text{m}^3$) of the Aethalometer to PAX ratio. The single scattering albedo was incremented from ~0.4 (100% Aquadag) to ~0.9 in steps of 0.1 in order to measure the Aethalometer to PAX ratio up to 80 ATN for each scattering ratio.

All target values were set using the PAX as the BC reference monitor assuming a mass absorption cross-section of 4.73 (m^2/g) (39), analogous to the attenuation cross section used to convert a measured attenuation change to a BC concentration in an Aethalometer. This operational definition of BC concentration is an important limitation of the Aethalometer (and other BC measurement techniques).

Ten-second data from the PAX and AE51 were smoothed using a Lowess algorithm (Igor Pro 6.3.7, Wavemetrics, OR, USA). The normalized BC_{AE51} to BC_{PAX} ratio (R_{meas}) was calculated from the smoothed data using Equation 1. Here, R_{meas} represents the change in AE51 response due to filter loading. Target ATN was achieved after 10 to 20 hours of sampling, resulting in approximately 5000 data points per instrument per experiment.

These experiments were performed using unloaded filters in up to three AE51s operating at nominal flows rates of 100 ml/min, 150 ml/min or 200 ml/min. Runs with AE51s at multiple flow-rates can be used to elucidate the effect of increased BC loading, because the change in attenuation will be less at a lower flow rate (due to the lower rate of PM mass accumulation on the filter) (40). Consequently, the divergence in the derived BC concentration between

these instruments will be due to the BC loading effect alone. Running AE51s at multiple flow rates will not elucidate the effects of scattering because ω will be the same.

2.5 Real-world correction factor validation

Chamber measurements were then conducted using previously loaded AE51 filters to assess the loading effect and correction equations for real-world measurements of personal BC exposure. Previously loaded AE51 filters were obtained from archived samples taken during the Fort Collins Commuter Study (35), a repeated measures panel study that collected 30-hour exposures for 46 volunteers over a two-year period. A total of 103 AE51 filters were retained from the Commuter Study for the following analysis; they were stored at -80°C and analyzed in batches. All participants provided written informed consent, and the Colorado State University Institutional Review Board approved all study procedures.

A target concentration of $2\text{ }\mu\text{g}/\text{m}^3$ of Aquadag was nebulized into the chamber. Pre-loaded filters taken from the Commuter Study were inserted into the same AE51 used for the exposure assessment, with the flow rate set to 200 ml/min. The instruments were then connected to the chamber via a low resistance HEPA filter (PALL Corp, NY, USA) for two minutes, ensuring the system was stable and leak free. The filter was then bypassed and aerosol was sampled onto the pre-loaded filters for five minutes. The AE51's flow rates were measured before and after each filter sample to validate their consistency. The PAX was zeroed before each sample (once connected to the chamber), with care to keep the inlet pressure stable until the end of the sample. This procedure was repeated for all 103 filter samples. Periodically (every 10th measurement), unloaded (clean) filters were inserted into the AE51.

The last four minutes of BC data from each five-minute sample were averaged. The pre-loaded filter samples provided BC_{Aeth} and BC_{Pax} values and the 'blank reference samples' provided $BC_{Aeth,Ref}$ and $BC_{Pax,Ref}$ values. Equation 1 was then used to calculate R_{meas} , the reduction in AE51 signal due to the loading of the filter during the Commuter Study.

2.6 Loading correction factors

Five different correction equations were applied to the AE51 measurements based on Coen et. al. (27), Drinovec et. al. (40), Kirchstetter and Novakov (41), Virkkula et. al. (13) and Weingartner et. al. (9) as shown in Table 1. The Drinovec, Virkkula and Kirchstetter equations use fit parameters derived from laboratory and field studies. The fit parameters in the Weingartner and Coen equations include the mean single scattering albedo ($\overline{\omega}$) of the sampled aerosol and a parameter related to the wavelength (λ) dependence. For these experiments $\overline{\omega}$ was calculated from the PAX. The Dr $\overline{\omega}$ inovec, Virkkula and Kirchstetter corrections were chosen because they have been most commonly applied to AE51 data. The Coen and Weingartner equations are more physically based (both relate to $\overline{\omega}$ and λ) and offer a more flexible solution to loading corrections; however, they have not been widely applied to AE51 data.

2.7 Correction equation optimization

Each correction equation was optimized by refitting the coefficients (a_i) to the real-world data set using Levenberg-Marquardt nonlinear least-squares fitting (42). The RMSE of the re-fitted equations was then compared to the original equations (using the previously published coefficients).

A new correction equation was also derived using the R statistical program (43, 44). The variables, their form and scaling were optimized using the R multivariable-fractional-polynomials (*mfp*) package (45, 46) that combines backwards-variable selection with polynomial transformations. At each elimination step, a detailed search of power transformation combinations is performed to determine whether a covariate should be added to the model and in what form. Terms judged to have improved the model are retained by *mfp* and the process of adding and removing variables is repeated until no changes to the output equation are made at each step. Our implementation of the *mfp* algorithm (code available upon request) used the deviance metric derived from the model maximum-log-likelihood at the $p=0.05$ level to accept or reject variables and their transformations in order of increasing complexity (45).

Two candidate variables, ATN and aerosol scattering at 880nm, were selected for inclusion in the new correction equation. The *mfp* analysis was performed on ATN and on $e^{-ATN/100}$ so that transformations analogous to all of the five existing transformations were considered. The aerosol scattering variable was derived from a portable nephelometer (pDR 1200, Thermo Inc.) run in parallel to the AE51 during the real-world exposure assessments (35). The pDR reports a mass concentration ($\mu\text{g}/\text{m}^3$), we averaged the concentration over the sample period and multiplied it by the sample duration to estimate the cumulative scattering.

3 Analysis and results

3.1 Response to Aquadag

Uncorrected, the Aethalometer underestimates BC levels by 16%, 31%, 42%, 51%, 59% and 69% at $ATN = 20, 40, 60, 80, 100$ and 125 respectively. This relationship between fractional reduction in AE51 response and ATN is illustrated for Aquadag aerosol in Figure 2, with the error in R due to the loading effect shown in the inset (black line). The PAX derived ω was 0.395 (S.D. = 0.013) for this experiment.

3.1.1 Standard coefficients—Application of correction coefficients from the literature (Table 2) results in Kirchstetter, Virkkula, and Weingartner equations under-predicting the loading effect. The performance of the five correction equations is also shown in Figure 2. The RMSE and MB associated with each correction equation are shown in Table 3. The Kirchstetter equation has the lowest RMSE (0.03) of all five corrections (Table 3). The RMSE is reduced slightly for the Kirchstetter, Virkkula and Weingartner equations when the ATN range is restricted to 0 and 80 (Table 3). The RMSE remains the same (within 1%) for the linear (Coen and Drinovec) equations when the ATN range is restricted to 0 and 80 (Table 3).

3.1.2 Optimized coefficients—When the coefficients for the correction equations are optimized specifically for Aquadag aerosol, the RMSE values are further reduced (Table 3). The Kirchstetter correction results in the smallest residuals across the measured attenuation range, with RMSE of < 0.01 . The Coen, Drinovec and Virkkula corrections results in residuals in the range of 0.07 to 0.09 at τ_{ATN} up to 100, above which they quickly increase. The form of the Weingartner equation produces large errors at τ_{ATN} less than ~ 10 , an issue acknowledged by the authors that the Coen equation is designed to rectify (27).

3.1.2 Response to NIST Diesel Particulate Matter—The loading effect of the NIST aerosol was the same as the Aquadag up to $\sim 60 \tau_{\text{ATN}}$. Above $60 \tau_{\text{ATN}}$ the NIST aerosol produced a loading effect increasingly larger than the Aquadag aerosol. At 70 and $80 \tau_{\text{ATN}}$ the NIST aerosol gave R values 0.02 and 0.05 lower than the Aquadag aerosol with respect with the normalized Aethalometer to PAX ratio.

3.1.3 Response to Aquadag – Ammonium Sulfate mixtures—No difference in the BC loading effect was observed from the accumulation of absorbing particles onto the filter as a function of τ_{ATN} as ω was increased from ~ 0.4 (pure Aquadag) to ~ 0.9 . The addition of ammonium sulfate results in a constant offset in the Aethalometer to PAX ratio. The normalization factor is dependent upon ω , but once applied the reduction in R_{meas} is the same function of τ_{ATN} .

3.1.4 Response to Ammonium Sulfate—The ammonium sulfate aerosol is almost entirely scattering as measured by the PAX ($\omega = 1.0$, S.D. = 0.01; $B_{\text{scat}} = 10.4 \text{ Mm}^{-1}$). The B_{CAE51} to B_{scat} ratio remained constant (0.005, S.D = 0.001) during these tests, demonstrating no cumulative loading effect from pure ammonium sulfate particles. The attenuation increased by only 2.3 across 24 hours with an AE51 sample flow of 200 ml/min. Consistent with other studies (47), the Aethalometer technique responds linearly to an increasing accumulation of scattering particles onto the filter and non-linearly to absorbing particles.

3.2 Real-world correction performance

One hundred and three loaded filters were analyzed. Two filters were removed from the analysis because they were overloaded ($\tau_{\text{ATN}} > 125$). The τ_{ATN} loadings on the remaining filters ranged from 7.6 to 112.4. Shown in Figure 3 (top panel, circles) are the reductions in concentration response, R , due to filter loading for each of the real-world filters. The loading effect was smaller than observed for the Aquadag aerosol. For the real-world samples R_{meas} was 0.1 higher on average, when compared to the Aquadag value at the same τ_{ATN} .

Performance of the five correction curves are shown in Figure 3 (top left panel) using the standard and fitted coefficients from Table 2. The other panels in Figure 3 depict the errors for each of the five correction equations using the standard fit coefficients. The trends in the errors highlight the increasing under-correction of the Coen, Virkkula and Weingartner equations and the increasing over-correction of the Kirchstetter and Drinovec equation.

3.2.1 Standard coefficients—The Drinovec and Kirchstetter correction tends to overcompensate the loading effect using the standard parameters, resulting in MB of -0.08

and -0.04 respectively compared to the reference concentration. Coen, Virkkula and Weingartner equations tended to undercompensate the loading effect (MB, +0.09, +0.05 and +0.08 respectively). The RMSE of the five equations ranged from 0.10 to 0.13 in the sub 125 ATN range.

Analysis of the standard correction factor residuals (illustrated in Figure 3, lower panel), showed a significant linear relationship between the error and attenuation for the Drinovec ($p < 0.05$), Weingartner ($p < 0.01$) and Coen corrections ($p < 0.01$). The Kirchstetter ($p = 0.3$) and Virkkula ($p = 0.1$) corrections did not show significant relationships between the error and attenuation.

3.2.2 Optimized coefficients—Re-fitting the five equations and restricting the ATN range to 80 reduced the RMSE to ~0.08 for all five equations (Table 3). The fitted ω in the Coen and Weingartner equations was in the range of 0.55 to 0.59. There was little difference in RMSE between the five optimized equations in the 5 to 60 ATN range where most of the real world data was collected.

3.3 The optimized correction equation for ATN less than 80

The *mfp* procedure did not suggest any variable transformations with both ATN and $(e^{-ATN/100})^{-1}$ as covariates. The $e^{-ATN/100}$ transformation, analogous to the Kirchstetter equation, resulted in the most normal residual distribution upon examination of the residual quantiles. However, the RMSE was 0.07 for both forms of the ATN covariate.

The *mfp* procedure suggested the cumulative scattering variable would improve the correction equation. The RMSE improved by 0.015 when the cumulative scattering variable was added (an improvement of 0.49 to 0.65 in the adjusted r-squared). Equation 6 shows the final form of the equation, where $\Sigma scat$ is scaled by 1×10^{-6} .

$$R_{eqn} = a_0 + a_1 \times e^{-\Delta ATN/100} + a_2 \times \sum scat + a_3 \quad (6)$$

where $a_1 = 0.833$ (standard error = 0.074), $a_2 = 0.078$ (standard error = 0.014) and $a_3 = 0.189$ (standard error = 0.053).

4 Discussion

The application of a loading correction is essential to remove bias from personal Aethalometer measurements; failure to do so means that data collected by these instruments is semi-quantitative at best. We have demonstrated that AE51 measurements will incur substantial negative bias due to the cumulative loading of BC during personal exposure assessment. With the attenuation range of the measurement limited to 80, loading corrections from the literature reduced the error to ~0.12 (RMSE). An informed choice of correction equation reduced the RMSE further to ~0.08 and removed correction equation dependent bias of $\pm 10\%$. For most AE51 applications there is probably only marginal benefit in improving the loading effect RMSE from 0.12 to 0.08, users are advised to use the

Coen, Drinovec, Kirchstetter or Virkkula equations with coefficients from the literature that best match their application.

The accumulation of both light absorbing (BC) and light scattering (non-BC) particles onto the AE51 filter over time will affect the AE51's ability to report BC concentrations accurately. Whereas the accumulation of purely scattering particles produces a linear effect on the estimated BC mass, the effect from absorbing particles (i.e., the accumulation of BC onto the filter) is highly non-linear.

The loading effect results in a systematic multiplicative error that increases as a function of ATN (48). The implications of not applying a loading correction will depend upon how the data is being used. Comparison of measurements made on filters with different loading levels will be subject to error, in the most extreme case ($R \sim 0.3$) a $20 \mu\text{g}/\text{m}^3$ measured on a fully loaded filter could actually represent an exposure in excess of $65 \mu\text{g}/\text{m}^3$. Studies where BC time-series are disaggregated, for example, as a function of job task or spatial location will suffer from this lack of inter-comparability due to the different loading levels prior to the period of interest.

The systematic multiplicative error due to the loading effect will also bias any BC exposure/response relationship (i.e., evaluation of a health outcome health) derived from AE51 data. The loading effect would result in lower exposures being associated with a given outcome than actually occurred. Public health policies based on the magnitude of BC exposure would be subject to error, if the loading effect were not accounted for.

The loading effect occurs across the entire operating range of the AE51 (from 0 to $125 ATN$) and the error from this effect would be impractical to avoid (either with frequent filter changes or by reducing the sample flow rate to minimize the attenuation change). The Aquadag experiments showed the loading effect increased more quickly at lower ATN , resulting in a 25% error at $25 ATN$ and a 50% error at $75 ATN$. Users should therefore apply one of the loading correction equations verified here, to account for the systematic error.

The user has several options to improve the accuracy of their BC measurements. First, collected AE51 data should be corrected using one of the aforementioned algorithms. Second, the user can optimize AE51 performance by minimizing the accumulation of BC on the filter while obtaining sufficient temporal resolution for their application. These conditions, depicted in Figure 4, are met through proper selection of sample duration, flow rate, and resolution (i.e., the elapsed time between BC readings), along with a priori knowledge of BC concentrations expected in the field. Areas in green delineate optimal operating conditions. In developing Figure 4, we adopted two important boundary conditions: that ATN should not exceed 80 during a measurement and that sample resolution be long enough to limit instrument noise. Hagler et al. (49) found that the measurement noise was minimized ($11 \text{ ng}/\text{m}^3$) for an attenuation change of 0.05 or greater. More recent studies (e.g. 20, 24) have found acceptable signal to noise ratios can be achieved over attenuation changes less than 0.05, perhaps as a result of improved AE51 firmware (e.g. flow control). As illustrated in the left panel of Figure 4, the AE51 can

provide BC concentration data at a resolution as fine as 60 seconds without exceeding 80 ATN ; such resolution is possible provided that: an optimal flow rate is selected, sample duration is less than approximately 24 hours, and the prevailing BC concentration is between ~ 1 and $10 \mu\text{g}/\text{m}^3$. Outside the aforementioned conditions, the user must choose a compromise between sampling duration and temporal resolution. The right panel of figure 4 illustrates how the AE51's sample resolution varies as a function averaging time for a range of BC concentrations.

Comparison of the NIST diesel particle standard and Aquadag suggests the AE51's ATN range should be limited to 80. The NIST standard and Aquadag samples start to diverge above $\sim 60 \text{ ATN}$, however the effect was small up to $\sim 80 \text{ ATN}$. For improved accuracy users could limit sampling to ATN values between 0 to 60, somewhat lower than what has been recommended elsewhere (e.g. 50) and well below the point (125) where the instrument reports its loading error. AE51 users could consider limiting the maximum ATN even further to minimize the size of the error they are correcting for. For example limiting ATN to 30% would limit the maximum error to 30% for aerosols with large loading effects similar to Aquadag (e.g. sooty urban aerosols).

The choice of optimal fit coefficients for a correction equation is challenging for personal exposure assessment. Even during fixed site monitoring where coefficients can be derived based on consecutive measurements on loaded and unload filters, the approach can breakdown in dynamic environments (27). For exposure assessment where the sampling is mobile and may move between distinctly different microenvironments this problem is likely exacerbated. Several options exist for reducing expert judgment in the coefficient selection including; a-priori knowledge of the sampling environment, concurrent scattering measurements (i.e., using a nephelometer in conjunction with the AE51), running two Aethalometers at different flow rates, and post analysis of the filter samples.

The loading correction coefficients from the present study may offer a better estimate for the overall loading in similar exposure scenarios. The loading correction coefficients from the present study may offer a better estimate for the overall loading in similar exposure scenarios. The "real-world" study 24-hour activity patterns comprised 14 hours (study median) at home, 7 hours at work, 1 hour in transit, less than 1 hour in eateries and 2 hours in other microenvironments in a mid-sized US city (51). The coefficients derived here are expected to reflect the mix of sources (local on-road traffic and cooking are likely to be two of the larger contributors in addition to longer range transport). Studies with a larger near-traffic contribution might expect coefficients closer to the Aquadag experiments. The extent to which the variability in particle composition influences AE51 measurements remains an active area of inquiry."

The current study focused mainly on bias due to BC loading, our ability to probe the effect of light scattering particles was limited. Also, while our method of normalizing the AE51 data to the PAX elucidates the loading effects it does not probe how changes in particle composition influence the relationship between ATN derived from the AE51 and the mass of BC. Our method provides a snapshot of the loading effect at the end of the sampling period. We cannot easily determine how the measurement error varies during the sample.

Ideally, the effect of aerosol scattering would be taken into account during sampling. Aerosol scattering measurements are often made in tandem with BC during exposure studies (e.g. 52, 53); such data might help further correct AE51 measurements. Cumulative scattering was a significant predictor of the AE51 to PAX ratio for the real world experiments, however its effect on the RMSE was fairly small.

The AE51 to PAX ratio however, was dependent on ω in the ammonium sulfate - Aquadag mixture experiments. Our results demonstrate the potential for including scattering in personal exposure studies, where sample size (i.e. lower cost instrumentation) is often a priority. More work is needed to understand the within sample variability of the scattering effect on BC estimates and how to optimally account for it as necessary in exposure assessment studies. The ω is expected to vary widely in different personal exposure situations. When exposure is dominated by fresh vehicle exhaust ω may be as low as 0.15 (41). When exposure occurs further away from primary sources, for example sub-urban settings, ω is likely to be much higher; 0.8 to 0.95 (41).

Running two Aethalometers at different flow rates can be used to quantify the magnitude of the BC loading effect. The downside of such an approach for personal exposure studies is cost (which would limit study sizes) and the additional burden on the study participant (which might modify behavior). A purpose-built, instrument with two measurements at different flow rates might alleviate one or both of these issues, however the added size and cost would likely prohibit deployment in many personal monitoring studies.

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Figure 1.
Schematic of the chamber and sampling setup

Figure 2.

Loading equations optimized to fit the 150 ml/min 10 $\mu\text{g}/\text{m}^3$ Aquadag experiment, up to 80 Δ_{ATN} . Main panel shows the measured fractional reduction in response (R_{meas}) with increasing attenuation derived from the normalized Aethalometer to PAX ratio (black line) alongside the reduction in response fitted using the five loading correction equations (R_{eqn}). The inset plot shows percent error of the five correction equations fitted with the standard coefficients (colored lines) and the error due to the uncorrected loading (black line).

Figure 3.

Real-world filter analysis. The upper panel shows the fractional reduction in response (R) with increasing attenuation after 36 hours of exposure assessment (circles) alongside the reduction in response from the five standard loading correction equations (colored lines). The measured response to Aquadag is shown for reference (dashed line). The five smaller panels show the errors of the five equations using the standard coefficients.

Figure 4.

Optimal Aethalometer (AE51) settings. Left panel depicts the maximum sample duration per filter as a function of black carbon (BC) concentration for each flow setting colored by the mean resolution per minute. Right panel depicts the AE51's resolution as a function of averaging time for mean BC concentrations between 0.1 and 1000 $\mu\text{g}/\text{m}^3$. Areas in green delineate optimal operating conditions.

Table 1

Correction equations. Where R is the fractional reduction in response, Δ_{ATN} is the change in percent attenuation, ω is the mean single scattering albedo and a_i are the fit coefficients.

Equation	Reference
$R = \left(\frac{1}{a_1 + 1} - 1 \right) \times \frac{\Delta_{ATN}}{50\%} + 1, \quad a_1 = a_1' \times (1 - \omega)$	Coen et al. 2010
$R = (1 - a_1 \times \Delta_{ATN}) a_1$	Drinovec et al. 2015
$R = a_1 \times \exp^{-\Delta_{ATN}/100} + a_2$	Kirchstetter and Novakov 2007
$R = \frac{1}{1 + (a_1 \times \Delta_{ATN})}$	Virkkula et al. 2007
$R = \left(\frac{1}{a_1} - 1 \right) \times \frac{\ln(\Delta_{ATN}) - \ln(10\%)}{\ln(50\%) - \ln(10\%)} , \quad a_1 = a_1' \times (1 - \omega) + 1$	Weingartner et al. 2003
$R = a_1 \times e^{-\Delta_{ATN}/100} + a_2 \times \sum scat + a_3$	This Work

Table 2

Fit coefficients (a_1') and mean single scattering albedos ($\overline{\omega}$) for the loading correction equations. The coefficients derived for attenuation changes less than 80 and less than 125 as well as the standard literature (standard) are shown.

Equation, ATN range	Aquadag	Real-world
Coen	0–80 $a_1' = 0.881, \overline{\omega} = 0.395^I$	$a_1' = 0.740^2, \overline{\omega} = 0.568$
	0–125 $a_1' = 0.714, \overline{\omega} = 0.395^I$	$a_1' = 0.740^2, \overline{\omega} = 0.563$
	Standard $a_1' = 0.740^2, \overline{\omega} = 0.395^2$	$a_1' = 0.740^2, \overline{\omega} = 0.750^2$
Drinovec	0–80 $a_1 = 0.007$	$a_1 = 0.005$
	0–125 $a_1 = 0.006$	$a_1 = 0.005$
	Standard $a_1 = 0.006$	$a_1 = 0.006$
Kirchstetter	0–80 $a_1 = 0.913, a_2 = 0.080$	$a_1 = 0.730, a_2 = 0.298$
	0–125 $a_1 = 0.944, b = 0.058$	$a_1 = 0.787, a_2 = 0.258$
	Standard $a_1 = 0.880^2, b = 0.120^2$	$a_1 = 0.880^2, a_2 = 0.120^2$
Virkkula	0–80 $a_1 = 0.012$	$a_1 = 0.007$
	0–125 $a_1 = 0.013$	$a_1 = 0.007$
	Standard $a_1 = 0.010^2$	$a_1 = 0.005^2$
Weingartner	0–80 $a_1' = 0.966, \overline{\omega} = 0.395^I$	$a_1' = 0.830^2, \overline{\omega} = 0.579$
	0–125 $a_1' = 1.122, \overline{\omega} = 0.395^I$	$a_1' = 0.830^2, \overline{\omega} = 0.552$
	Standard $a_1' = 0.830^2, \overline{\omega} = 0.395^I$	$a_1' = 0.830^2, \overline{\omega} = 0.750^2$
This work	0–80 NA	$a_1 = 0.833, a_2 = 0.078, a_3 = 0.189$

^I Measured

² Held constant

Mean bias (MB), root mean square error (RMSE) and sample size for the 10 µg/m³ Aquadag and real-world data for attenuation changes () ATN) up 80 and 125 using the fitted and standard fit coefficients.

Table 3

Equation, coefficients, ATN range		Aquadag (n = 3275 ^I)		Real-world (n = 103 ²)	
		MB	RMSE	MB	RMSE
Coen	Standard Coefficients	0–125	0	0.05	0.13
		0–80	0.04	0.08	0.12
	Fitted Coefficients	0–125	NA	NA	0.09
		0–80	NA	NA	0.08
Drinovec	Standard Coefficients	0–125	0.02	0.05	0.10
		0–80	0.05	–0.03	0.09
	Fitted Coefficients	0–125	NA	NA	0.09
		0–80	NA	NA	0.08
Kirchstetter	Standard Coefficients	0–125	0.03	0.03	0.12
		0–80	0.02	–0.08	0.11
	Fitted Coefficients	0–125	NA	NA	0.09
		0–80	NA	NA	0.08
Virkkula	Standard Coefficients	0–125	0.06	0.08	0.11
		0–80	0.03	0.04	0.10
	Fitted Coefficients	0–125	NA	NA	0.09
		0–80	NA	NA	0.08
Weingartner	Standard Coefficients	0–125	0.09	0.10	0.13
		0–80	0.06	0.07	0.11
	Fitted Coefficients	0–125	NA	NA	0.09
		0–80	NA	NA	0.08
This Work	Fitted Coefficients	0–125	NA		NA
		0–80			0.07

Note: MB not applicable to fitted data

^INumber of data points in the 0–125 ATN fit

Number of filter samples analyzed

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