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Loading Effect Correction for Real-Time Aethalometer Measurements of Fresh Diesel Soot

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ABSTRACT

In this study, a correction was developed for the aethalometer to measure real-time black carbon (BC) concentrations in an environment dominated by fresh diesel soot. The relationship between the actual mass-specific absorption coefficient for BC and the BC-dependent attenuation coefficients was determined from experiments conducted in a diesel exposure chamber that provided constant concentrations of fine particulate matter (PM; $PM_{2.5}$; PM <2.5 μm in aerodynamic diameter) from diesel exhaust. The aethalometer reported BC concentrations decreasing with time from 48.1 to 31.5 $\mu g\ m^{-3}$ when exposed to constant $PM_{2.5}$ concentrations of 55 ± 1 $\mu g\ m^{-3}$ and b_{scat} = 95 ± 3 Mm^{-1} from diesel exhaust. This apparent decrease in reported light-absorbing PM concentration was used to derive a correction *K*(*ATN*) for loading of strong light-absorbing particles onto or into the aethalometer filter tape, which was a function of attenuation of light at 880 nm by the embedded particles.

INTRODUCTION

Carbonaceous aerosols are composed mainly of organic carbon (OC) and elemental carbon (EC), the latter a measure of black carbon (BC) aerosol. Carbonaceous aerosols may have a detrimental impact on human health from both acute and chronic exposures because of their toxicological effect and because they are able to penetrate into the lower respiratory system.[1,2] Susceptible populations include those suffering from chronic obstructive pulmonary disease, cardiovascular patients, and children with asthma.[3–6] In addition, BC aerosol influences climate directly and indirectly through light extinction in the atmosphere[7–9] and can lead to low visibility.[10,11]

It is known that the absorption efficiency of BC aerosol varies depending on the source and chemical composition,[12–16] and the assumption that all light-absorbing material is because of the presence of BC aerosol is not always accurate.[17–21] Light-absorption efficiencies of aerosols embedded in a reflective filter matrix are known to be enhanced compared with the same aerosols in the atmosphere.[22–25] For an early version of the aethalometer operating with an incandescent lamp, a value of σ_{ATN} = 19 $m^2\ g^{-1}$ was used to translate changes in light attenuation through a quartz filter into BC mass concentration.[26] This value was compared with the absorption efficiency of similar aerosols in the air (σ_{abs}~10 $m^2\ g^{-1}$; Horvath[8]) to account for absorption enhancements because of multiple scattering within the filter matrix.[23,24]

In addition, an optical effect because of the accumulation of particles in the filter has been reported for the aethalometer[25,27–29] and similar applications of the light transmission method.[13,30] As the filter becomes loaded with particles, the enhancement of the light absorbed per unit mass of added BC decreases, which results in lower reported BC concentrations for loaded filters compared with lightly loaded filters. This optical effect is referred to as a "shadowing effect." Weingartner et al.[28] reported that this effect is more pronounced for freshly emitted soot than for aged atmospheric aerosol. The current aethalometer algorithm used to translate filter light attenuation into BC mass concentration does not correct for this loading effect. In this study, we examine this optical effect using a constant source

IMPLICATIONS

The aethalometer is used to measure near real-time concentrations of light-absorbing aerosols, but may not capture the real temporal behavior of airborne diesel soot concentrations because of variability in measured optical attenuation related to loading of the filter with strong light-absorbing particles. To examine short-term average concentrations of diesel soot, the instrument requires a correction for mass buildup of high light-absorbing particles on the filter. This is because the amount of the light absorbed per unit of BC mass decreases, which results in lower reported BC concentrations for loaded filters compared with lightly loaded filters.

of diesel soot, and a correction for the current aethalometer algorithm was developed to measure real-time concentrations of BC from fresh diesel exhaust.

EXPERIMENTAL WORK

Two portable versions of the dual-wavelength aethalometers (AE41 and AE42, Magee Scientific Company) were simultaneously exposed to controlled concentrations of ultrafine diesel-generated particles (soot) in a specially designed diesel chamber. We used two exposures at variable concentrations of fine particulate matter (PM; $PM_{2.5}$) and one at a constant (55 ± 1 μg m^{-3}) concentration of $PM_{2.5}$ from diesel soot, which was used to derive the correction to the aethalometer algorithm. The conditions of approximately 50 μg m^{-3} of $PM_{2.5}$ from diesel exhaust generated for the experiments were comparable to similar pollution observed near a high traffic road in Paris, France,[31] and lower than the BC concentrations observed when chasing a transit bus in Los Angeles, CA.[32]

Diesel Chamber

The diesel chamber was located in Seattle and was operated by the Department of Environmental and Occupational Health Sciences at the University of Washington. The chamber dimensions were 8.8 × 5.5 × 2.4 m with a total volume of 116 m^3. The volumetric flow rate through the chamber was 28.3 m^3 min^{-1}, and the incoming air was filtered so that the PM concentration inside the chamber was unaffected by background PM. Diesel soot was generated from a turbocharged direct-injection 5.9-L Cummins B-series diesel engine (6BT5.9G6, Cummins, Inc), which was comparable to engines used in delivery trucks and school buses. The engine drove a 100-kW generator connected to an electric load bank (Simplex), and the load applied to the running engine was set to 75 kW. The engine fuel was highway grade diesel No. 2 undyed, which is commonly used in delivery vehicles.

The $PM_{2.5}$ concentration inside the chamber was established by a two-stage dilution process that mixed air with diesel exhaust. The degree of dilution and resulting concentration was adjusted with a variable speed fan that was electronically controlled by a system that used two light-scattering nephelometers (one sensing upstream of the chamber and the other inside the chamber). This provided feedback to the system to adjust the amount of diverted diesel exhaust to achieve and maintain a target $PM_{2.5}$ concentration. Under these conditions of controlled constant $PM_{2.5}$ concentration from diesel exhaust, the chamber exhibited a linear relationship between EC and $PM_{2.5}$ mass concentration (intercept = −6.1; slope = 0.85; R^2 = 0.97).

Measurements

To verify constant conditions and to compare chamber-generated diesel soot to other experiments reported in the literature, several supporting PM parameters were measured in the chamber, including continuous $PM_{2.5}$ concentration from a tapered element oscillating microbalance (TEOM) monitor with a $PM_{2.5}$ inlet (1-min; Series 1400a, Thermo Electron Co.) and light scattering coefficient, b_{scat}, from a nephelometer (1 min; M903, Radiance Research). The nephelometer was calibrated using zero air and carbon dioxide for span setting. In addition, integrated $PM_{2.5}$ samples were taken from collocated single-stage 5 L min^{-1} low-volume samplers (Airmetrics Inc.). Two-stage filter cassettes (47-mm) with Teflon filter (2-μm pores; part 7592-104, Whatman Inc.) and quartz filter after Teflon, as well as single-stage filter cassettes with quartz (part 1851047, Whatman Inc.), were used with the samplers to estimate OC and EC fractions of the $PM_{2.5}$. In addition, two EcoChem samplers (EcoChem Analytics) were deployed to measure particle active surface area (PASA) and total particle-bound polycyclic aromatic hydrocarbons (PPAHs). These instruments have been used to characterize sources and type of aerosols based on the relationship between PPAHs and particle active surface area.[33,34]

Sample and Data Analysis

$PM_{2.5}$ collected on the Teflon filters was analyzed gravimetrically using a microbalance (model UMT2, Mettler-Toledo, Inc.) at a constant temperature (22.2 ± 1.8 °C) and relative humidity (34.8 ± 2.5%), after the samples were equilibrated for ≥24 hr before weighing. Sections of the quartz filters (1.5 cm^2) were analyzed for OC and EC via thermal optical evolved gas analysis (thermal optical transmittance [TOT], Sunset Laboratory Inc.) using a modified version of the National Institute for Occupational Safety and Health 5040 method.[33] The TOT carbon analyzer was calibrated using a standard solution of sucrose (20 μL of 4.5 g C L^{-1} solution = 90 μg C). More details about the temperature steps, carrier gases, and standards used in this thermal optical analysis are reported in Pang et al.[35]

BC concentrations from the aethalometer were compared with the EC concentrations measured by thermal optical analysis of the quartz filters. In addition, aethalometer measurements were compared with other PM measurements, including $PM_{2.5}$ concentration, b_{scat}, PASA, and PPAHs. This was done to evaluate the temporal behavior of the aethalometer measurements during the chamber experiment at constant PM concentrations. Finally, a correction was proposed for the aethalometer to account for loading effect of the filter-particle matrix.

Model Framework

This study developed a correction for the aethalometer algorithm that could account for the loading effect. This work is specific for air with fresh diesel exhaust dominated aerosols.

The optical attenuation (ATN) of light by particles deposited in the quartz filter is given by the following relationship:

$$ATN = -100 \times \ln(I/I_0) = -100 \times \ln(T) \quad (1)$$

where I_0 is the light intensity through the unloaded filter media, I is the light intensity remaining after passing through the particles collected on the filter, and T is transmission. Aethalometers measure light attenuation through a quartz fiber matrix, which has high light scattering capabilities, so the light scattered away from the detector by the particles is assumed negligible

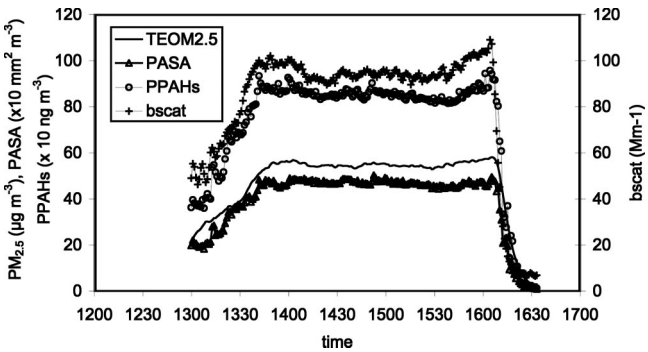

Figure 1. Temporal distribution of $PM_{2.5}$ concentration, PASA, PPAHs, absorption coefficient (b_{abs}), and scattering coefficient (b_{scat}) during exposure to diesel soot. Note constant levels of $PM_{2.5}$ between 2:00 and 4:00 p.m.

compared with the scattering by the filter.[26] The attenuation coefficient, b_{ATN}, has units of m^{-1} and is given as follows:

$$b_{ATN} = \frac{A}{Q}\frac{\Delta ATN}{\Delta t} \quad (2)$$

where A is the filter collection area (m_2), or "spot", Q is the volumetric flow rate (m^3 min^{-1}), and ΔATN is the change in light attenuation during the time interval, Δt (min). The mass concentration of BC aerosol, *MBC* (g m^{-3}), is then calculated from the attenuation coefficient:

$$M_{BC} = \frac{b_{ATN}}{\sigma_{ATN}} \quad (3)$$

where σ_{ATN} is the attenuation efficiency of BC in the filter substrate, with a manufacturer's recommended value of $\sigma_{ATN} = 16.6$ m^2 g^{-1} at $\lambda = 880$ nm and 39.5 m^2 g^{-1} at $\lambda = 370$ nm for the dual wavelength aethalometer, based on the relationship between attenuation and BC concentration determined by thermal optical analysis.[36]

The light absorption by the particles can be related to the total light attenuation (filter plus particles) by accounting for particle loading effect on the filter matrix with the correction factor, *K*(*ATN*), which accounts for a nonlinear absorption response to filter loading, as follows:

$$M_{BC} = \frac{b_{ATN}}{\sigma_{ATN}} \times 1/K(ATN) \quad (4)$$

and

$$K(ATN) = \frac{\Delta ATN(t)}{\Delta ATN(0)} \quad (5)$$

where relative increment in attenuation in a Δt interval, ΔATN(t), is the difference between two consecutive measurements [i.e. $ATN(t+1) - ATN(t)$], and $\Delta ATN(0)$ is the difference between the first and second reported measurements for light attenuation at the beginning of the measuring cycle of the instrument (new filter spot). Ideally, if the concentration of BC remains constant, the $\Delta ATN(t)$ measured on the filter at any given time, t, should remain constant and $K(ATN) = 1$. However, $\Delta ATN(t)$ progressively decreases with time during the measuring cycle because of the loading effect identified in this work.

In this study, we calculated [*K*(*ATN*)] for diesel soot based on 1-min averaged aethalometer measurements collected during a 2-hr period of constant concentrations of diesel exhaust particle mass. The experimentally derived *K*(*ATN*) was described as follows:

$$K(ATN) = a + b \times \exp(-\mathrm{ATN}/100) = a + b \times T \quad (6)$$

where a and b are linear regression coefficients.

RESULTS AND DISCUSSION

Diesel Chamber Experiments

TEOM $PM_{2.5}$, nephelometer scattering coefficient (b_{scat}), PASA, and PPAH concentrations in the diesel chamber experiment at constant $PM_{2.5}$ concentrations are shown in Figure 1. This figure shows the temporal evolution of the $PM_{2.5}$ concentration starting from and increasing in trend when diesel exhaust was mixed with filtered air until reaching a target concentration and a decaying trend at the end of the experiment when the engine was turned off. The time period of constant $PM_{2.5}$ concentrations between 2:00 p.m. and 4:00 p.m. (2 hr) was selected to test the aethalometer response to constant BC mass concentration and to determine a correction factor, *K*(*ATN*), to account for loading effect. The PM parameters measured during the 2-hr run at constant $PM_{2.5}$ concentration were 95 ± 3 Mm^{-1}, 470 ± 11 mm^2 m^{-3}, and 856 ± 23 ng m^{-3} for b_{scat}, PASA, and PPAHs, respectively. Note that all of the parameters were stable (~3% variation) and highly correlated ($r > 0.87$) to each other (see Table 1). We found, for this experiment, that the ratio of PPAHs/PASA was

Table 1. Summary of Spearman's correlation coefficients (N = 200) among TEOM $PM_{2.5}$, b_{scat}, PASA, and PPAHs for the diesel chamber experiment.

Variable	$TEOM_{2.5}$	b_{scat}	PASA	PPAHs	b_{ATN}/K 880 nm	b_{ATN}/K 370 nm
TEOM $PM_{2.5}$	1				0.94	0.91
b_{scat}	0.94	1			0.97	0.97
PASA	0.98	0.96	1		0.96	0.94
PPAHs	0.98	0.97	0.99	1	0.97	0.95
b_{ATN} 880 nm	0.87	0.93	0.90	0.91	–	
b_{ATN} 370 nm	0.84	0.92	0.87	0.89		–

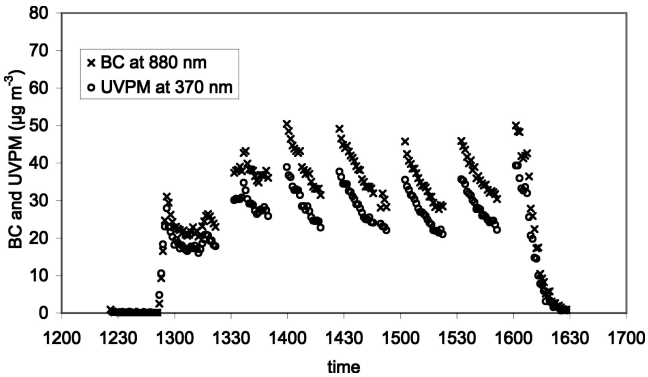

Figure 2. Temporal distribution of BC and UVPM aethalometer measurements during diesel soot exposure at constant levels of $PM_{2.5}$ from diesel exhaust.

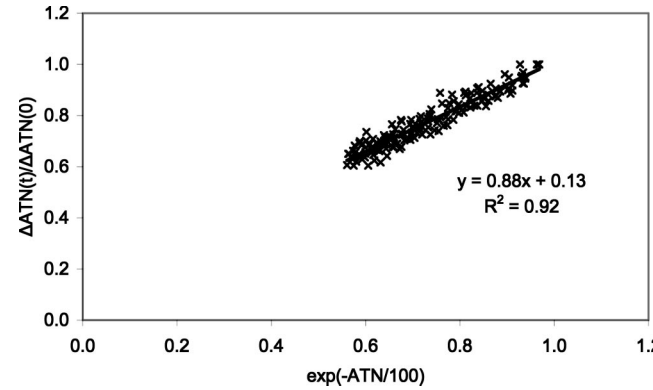

Figure 4. Linear regression applied to determine the regression coefficients *a* and *b* at 880 nm used to correct for particle loading effect in the aethalometer measurements.

1.8 ± 0.1 ng mm^{-2}, higher than similar ratios reported by Velasco et al.[33] from measurements in a road near a working dump truck (~1.2 ng mm^{-2}) and averaged measurements (~1 ng mm^{-2}) reported from chasing a tractor-trailer truck on a California road.[34] This suggests that, for this experiment, the diesel soot had more polycyclic aromatic hydrocarbons (semivolatile organic compound) attached to the particle surface in proportion to the available surface area than other reported diesel exhaust PM.

The aethalometer measurements obtained during the same diesel chamber experiment are shown in Figure 2. This figure includes uncorrected measurements for both channels (BC at 880 nm and UVPM [UV light absorbing PM] at 370 nm). Note the recurring decrease in the aethalometer measurements of BC and UVPM concentrations at constant $PM_{2.5}$ concentrations from diluted diesel exhaust. This periodic behavior was observed in all of the measurement cycles (four spots) and in both collocated aethalometers (sampling at 2.2 L min^{-1}). This is also evident in Figure 3, where the reported (cumulative) *ATN* values increased continuously, but not linearly, up to approximately 55 at 880 nm and 95 at 370 nm. The decreasing trend in the temporal evolution of the aethalometer measurements (from 48.1 to 31.5 μg m^{-3}; see Figure 2) at constant $PM_{2.5}$ concentrations suggests that the instrument underestimates BC concentrations when the filter is loaded with diesel soot. In addition, note from Figure 2 an apparent UVPM < BC signal (ratio UVPM/BC: ~0.75) in the reported measurements. Ideally, the BC concentration (determined from absorption at 880 nm) should be about equal to the UVPM concentration (determined at 370 nm) because of the inverse relationship between light absorption and wavelength for a blackbody. However, the difference in the reported concentration of light-absorbing PM from the two available channels results from the instrument's preset attenuation efficiency.

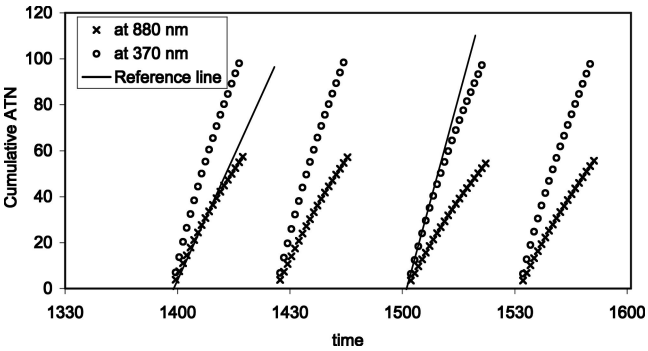

Figure 3. Cumulative ATN reported by the aethalometer at 880 and 370 nm during the experiment at constant $PM_{2.5}$ concentrations from diesel exhaust.

Aethalometer Correction

Values for *K*(*ATN*) were determined using the relationship presented in eq 5 for aethalometer measurements (N = 170) at constant concentrations of diesel-generated PM. Linear regression of *K*(*ATN*) versus *T* or exp(−*ATN*/100) produced the coefficients $a = 0.13$ and $b = 0.88$ (R^2 = 0.92) at 880 nm, in eq 6 (see Figure 4). Nearly identical values of regression coefficients were reported by Kirchstetter et al.,[29] who applied a similarly strong light-absorbing BC (i.e., with a single scattering albedo of ~0.25) from a diffusion flame to develop a correction for real-time aethalometer data. The agreement between the two approaches suggests that the coefficients reported here can

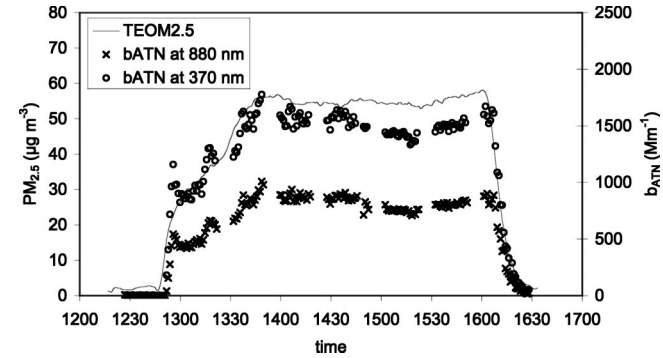

Figure 5. Correction for particle loading effect *K*(*ATN*) applied to the attenuation coefficient determined with the aethalometer. Note that the b_{ATN} measured at both wavelengths matches the temporal behavior of the $PM_{2.5}$ concentration.

be used when using the aethalometer to sample aerosol characterized by low single-scattering albedo in addition to diesel soot.

A similar approach was used for correcting the attenuation measurements at 370 nm (UVPM), which gave the coefficients $a = 0.38$ and $b = 0.67$ ($R^2 = 0.95$). The corrected b_{ATN} using the $K(ATN)$ correction for filter loading are shown in Figure 5 [corrected value = $b_{ATN}/K(ATN)$]. There is good agreement between the temporal evolution of the $PM_{2.5}$ concentration and the corrected b_{ATN} measured with the aethalometer. In addition, after applying the above correction, the Spearman's correlation coefficient, r (Table 1), improved (>4% points) for all of the measured PM parameters.

CONCLUSIONS

We have developed a correction for the aethalometer measurement that enables reliable measurement of real-time concentrations of BC in an environment dominated by fresh diesel soot. This included a correction for a nonlinear response of the instrument as the filter becomes loaded with light-absorbing particles. This correction function was $K(ATN) = 0.38 + 0.67 \times \exp(-ATN/100)$ for attenuation of light measured at 880 nm and can be applied, in addition to diesel soot, when measuring BC in environments dominated by aerosols with low single-scattering albedo. We used a specially designed chamber for developing this correction because of its capabilities to actively maintain constant concentrations of $PM_{2.5}$ from diesel exhaust emitted from an engine operating under load conditions. This was necessary to determine the optical artifact occurring as the filter loads with particles. Additional work should include determining the attenuation efficiency of soot from fresh diesel exhaust. More work is required for testing the proposed correction with mixed aerosol, including nonabsorbing aerosols, as well as in ambient air with biomass smoke, which has an important component of semivolatile OC that may affect the light absorption measurement from adsorption onto the aethalometer's quartz filter.

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