

Comparison of Measurement Strategies for Light Absorbing Aerosols from Modern Diesel Engines

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ABSTRACT

Light absorbing components of aerosols, often called black carbon (BC), are emitted from combustion sources and are believed to play a considerable role in direct atmospheric radiative forcing by a number of climate scientists. In addition, it has been shown that BC is associated with adverse health effects in a number of epidemiological studies. Although the optical properties (both absorbing and scattering) of combustion aerosols are needed in order to accurately assess the impact of emissions on radiative forcing, many models use radiative properties of diesel particulate matter that were determined over two decades ago. In response to concerns of the human health impacts of particulate matter (PM), regulatory bodies around the world have significantly tightened PM emission limits for diesel engines. These requirements have resulted in considerable changes in engine technology requiring updated BC measurements from modern engines equipped with aftertreatment systems. In this study, a variety of common ambient monitoring techniques were used to characterize the light absorbing properties of diesel aerosol. Aerosol optical properties were directly measured with an Aethalometer and Photoacoustic Extinctionmeter and compared to filter based analysis. The results showed excellent correlation ($R^2 = 0.95$) between aerosol light absorption at the short IR wavelength with elemental carbon (EC) concentration from a thermal optical reflectance, NIOSH 5040 method. Resulting EC mass absorption cross-section efficiencies differed by 25 to 30% from manufacturer published values indicating the optical properties used by the instrument may not be representative of modern diesel engine emissions.

CITATION: Robinson, M., Liu, Z., Olson, M., and Schauer, J., "Comparison of Measurement Strategies for Light Absorbing Aerosols from Modern Diesel Engines," *SAE Int. J. Fuels Lubr.* 7(2):2014, doi:10.4271/2014-01-1570.

INTRODUCTION

Vehicular emissions are a prominent source of particles found in urban environments [1]. Particulate matter (PM), although remains the only regulated criteria pollutant that is not chemically defined, is one of the most important due to its adverse impact on human health, atmospheric conditions, and climate. A multitude of epidemiological studies have shown an association between ambient PM and harmful cardiovascular and pulmonary health effects [2, 3]. In addition to the health concerns related to PM, the optical behavior of a given aerosol is of growing concern due to its effect on atmospheric radiative forcing [4]. In response to these concerns, worldwide regulatory bodies have significantly tightened particulate emission limits for light and heavy duty vehicles. Since 2007 in the US, on-highway diesel engines have been using diesel particulate filters (DPFs) to meet PM mass emission standards. However, many off-highway applications are able to meet PM

mass emissions standards without a DPF, only using a diesel oxidation catalyst (DOC) and selective reduction catalyst (SCR). The application of modern PM control strategies significantly changes the chemical, physical and optical properties of diesel PM [5]. These changes to the makeup of emitted diesel PM require updated measurement methods to understand the possible environmental impact of control strategies.

Atmospheric modeling has shown that PM mitigation strategies have the potential of reducing mean projected global warming by $\sim 0.5^\circ\text{C}$ by 2050 [6]. Carbonaceous PM can be divided into Black Carbon (BC) and Organic Carbon (OC), where BC has been defined by the U.S. Environmental Protection Agency (EPA) as "a solid form of mostly pure carbon that absorbs solar radiation (light) at all wavelengths." [7]. BC is not the only constituent of PM which affects the radiative forcing potential of

an aerosol. A coating of OC has been shown to enhance light absorption at all wavelengths; this phenomena can be explained by the OC behaving like a lens, focusing the incident radiation into the BC core [4]. The term "Brown Carbon" (BrC) has been applied to the absorption of light by OC and is thought to be significant in overall aerosol light absorption measurements. This study aimed to reduce the impact of BrC on absorption measurements, as BrC measurements are still under development and add considerable uncertainty to light absorption.

Controlling BC emission has the near term benefit of reduced global mean radiative forcing as compared to other Green House Gases (GHGs) due to their reduced residence time in the atmosphere. Atmospheric BC concentrations have been measured across the country and the world for over 20 years. In the US, over half of BC emissions in 2005 were due to mobile sources, in which nearly 90% were attributed to diesel engines [7]. Measurements in California have shown the effect of reduction of diesel PM emissions on atmospheric BC concentrations [8]. The determination of optical properties of the aerosol is crucial for the accuracy of radiative transfer modeling, unfortunately many models use values over two decades old [4]. One of the aims of this work is to compare modern diesel engine BC to historical diesel BC.

Due to increasingly stringent air quality regulations, a large amount of measurement techniques have been developed to quantify aerosol light absorption. These methods have been summarized extensively by Moosmüller [9]. Some of the methods of interest, when regarding diesel engine source testing, include thermal optical reflectance (or transmittance) analysis of quartz particulate matter filters (Elemental Carbon/Organic Carbon), in situ photoacoustic light absorption, and real time quartz filter tape transmission (Aethalometer). Integrated filter based methods are widely accepted but lack time resolution required for transient diesel emissions and have long turnaround time compared to real time instruments [9]. Real time instruments have been developed to correlate with integrated filter measurements (most often EC) [10]. Extensive work has been done characterizing and describing all aforementioned methods and is out of scope of this paper.

The use of aerosol light absorption measurements in a heavy duty diesel emissions lab has been limited. Many resources have been devoted to measuring aged light absorption of atmospheric aerosols [4, 11, 12, 13, 14]. One of the first attempts of light absorption measurement of freshly emitted diesel aerosol from a Constant Volume Sampling (CVS) emissions measurement system was done by Moosmüller et al in 2001. Researchers struggled to measure BC emissions with the aethalometer due to dilution system constraints [15].

Kittelson et al. provided a comparison of carbon measurements using an aethalometer, NIOSH Method 5040 EC/OC integrated filters and a Scanning Mobility Particle Sizer [16]. The authors state, 'Since the aethalometer software uses a constant extinction cross-section value ($16.6 \text{ m}^2\text{g}^{-1}$) to calculate BC mass concentration, the importance of field calibration to determine a unique sigma value for various aerosols should be stressed.' [16]. Over the course of this literature review this sentiment of highly uncertain aerosol absorption cross-section efficiency (or sigma) for freshly emitted diesel particulate was common. An analytical solution for the mass specific absorption efficiency gives a framework for understanding BC measurement sensitivity.

Absorption efficiency is a function of particle size (D_p), the wavelength of the incident light (λ) and the complex refractive index of the particle (m). Eq. (1) describes the mass specific absorption efficiency of a smooth homogeneous particle.

$$E_{abs}(D_p, \lambda, m) = \frac{3}{2\rho_p D_p} Q_{abs}(m, \alpha) \quad (1)$$

Where ρ_p is the particle density, Q_{abs} is the dimensionless absorption efficiency and α is the dimensionless size parameter. Liu et al. offer a comparison of three size resolved particle density corrections [17]. An optical model was run with constant particle density, but varied over the expected range (0.2 g/cc to 1.2 g/cc) to demonstrate the sensitivity of optical properties as a function of particle composition. A numerical solution for Q_{abs} was provided by Bohren and Huffman [18, 19, 20]. Using a MATLAB based Mie theory model, the mass specific absorption efficiency were calculated using a refractive index of $1.68 + 0.56i$ from Bond and Bergstrom [4]. The results are presented in Figure 1.

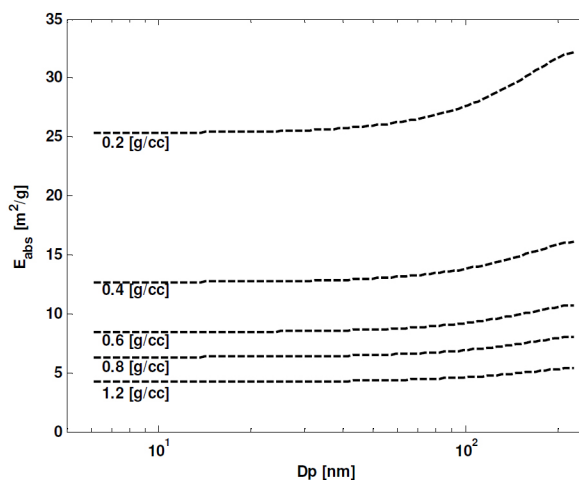


Figure 1. Mass specific particle absorption efficiency for diesel soot

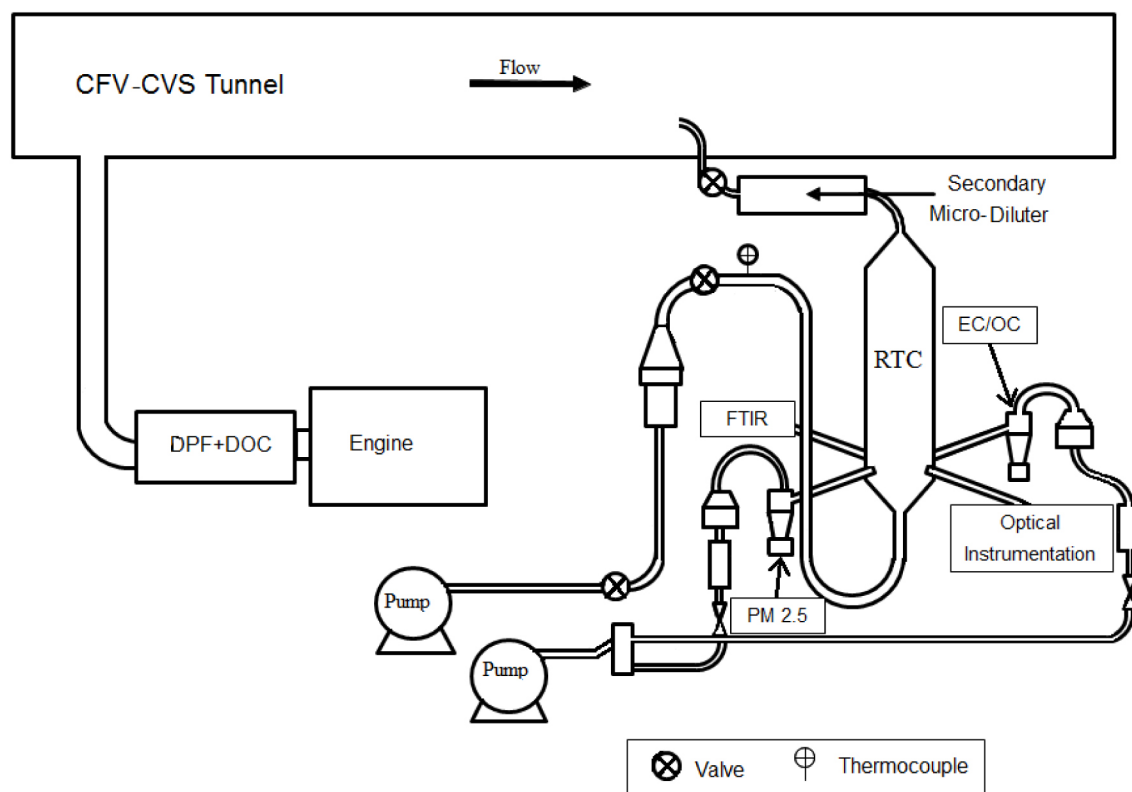


Figure 2. Test Cell and Dilution System Schematic

Figure 1 shows that mass specific absorption efficiency at small particle sizes (< 50 nm) with a constant particle density does not change strongly with particle size. However above 50 nm, mass specific absorption efficiency for a constant particle density can increase by 15% or greater. At a constant particle diameter, the range of expected particle densities can change mass specific absorption efficiency by a factor of 5. This necessitates particle size and composition information for fundamental understanding of diesel PM absorption measurements.

Modern diesel engine emission control strategies are known to change PM composition and particle size distribution (PSD) [5]. Post DPF optical measurements will likely lead to similar conclusions of PM mass reduction (greater than 99% filtration efficiency). However, a large fraction of diesel engines that operate off-highway, are able to meet Tier IV Final regulations without a DPF. This raises the concern of high BC emission rates for engines only equipped with a DOC.

The goal of the present study was to perform a comparison of the two available direct optical measurements in parallel with filter based methods. In order to gain a more fundamental understanding of factors affecting aerosol optical properties, particle size measurements were also done.

EXPERIMENTAL METHODS

Engine Setup

A turbocharged inline 6 cylinder heavy duty diesel engine was tested in this study. This engine uses high pressure common rail direct fuel injection, high pressure exhaust gas recirculation (EGR) and other common combustion strategies used to meet EPA 2010 emissions regulations. A DOC was used to reduce Carbon Monoxide (CO), Total Hydrocarbons (THCs) and some of the soluble organic fraction (SOF) of PM. It must be noted that during this study no selective catalytic reduction (SCR) system was used to reduce nitrogen oxides (NO_x), as the effect of SCR on PM optical properties was deemed out of scope. A General Electric 700 HP transient DC dynamometer was used to load the engine over three engine modes described in Table 1. Ultra low sulfur #2 diesel fuel was used during this study. Fuel specifications can be found in Table 2.

Table 1. Engine Operating Conditions

	Idle	B50	C100
Speed	650 RPM	1555 RPM	1912 RPM
Load	10 %	50 %	100 %
Exhaust Temp.	128 °C	341 °C	424 °C