Real-Time Estimation of Elemental Carbon Emitted from a Diesel Engine

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New Mining Safety and Health Administration (MSHA) regulations limit the mass concentration of airborne diesel particulate matter (DPM) or, more specifically, the concentration of elemental carbon (EC), in underground mines. The mine operators are responding by introducing a variety of controls to reduce DPM in the mines, potentially including the evaluation of new maintenance procedures to reduce underground mine vehicle emissions. There is currently a lack of an inexpensive and dependable method to directly measure the DPM concentration emitted from the vehicle tailpipe. To that end, this work demonstrated a simple field portable method for estimating the mass concentration of elemental carbon exiting the tailpipe of a diesel engine using a direct reading photometer. Simultaneous measurements of tailpipe exhaust were made with a Thermo Electron Personal DataRAM 1200 photometer (particulate mass concentration based on light scattering) and by analyzing $PM_{2.5}$ and $PM_{1.0}$ samples collected on quartz fiber filters using the National Institute of Occupational Safety and Health (NIOSH) method 5040 (mass concentration of EC via thermal−optical method). Results indicate surprisingly good correlation ($R^2 = 0.97$) of the two methods when the data are adjusted for relative humidity (RH) and corrected using an empirically generated calibration factor. Although preliminary, it may be possible to implement this method in maintenance shops to monitor emission trends and to compare emissions of various vehicles in a fleet. Such data will be useful for fleet planning to meet new air quality standards.

1. Introduction

New Mining Safety and Health Administration (MSHA) regulations limit the mass concentration of airborne diesel particulate matter (DPM) or, more specifically, the concentration of elemental carbon (EC), in underground mines (*1*). Mine operators have responded by introducing a variety of controls to reduce DPM in the mines (*2*−*4*). One control used by some operators is to implement more rigorous vehicle maintenance procedures. Although methods have been developed to evaluate a variety of engine emissions for maintenance purposes (*5*−*7*), there is still a need for an inexpensive and dependable method to directly measure the EC concentration at the vehicle tailpipe.

When detailed measurement of size distribution of the engine-emitted particulate matter is required, instruments such as the scanning mobility particle sizer (SMPS) or the real-time engine emissions particle sizer (EEPS) instrument (made by TSI Inc[1]) may be used. (The use of company names in this document does not imply NIOSH endorsement of any company or product). Such instruments are unfortunately relatively expensive, usually require dilution of the exhaust aerosol, and are generally not well-suited for use in an underground diesel maintenance shop due to their unique operating requirements such as accurate control and measurement of dilution air. In addition, although particulate mass can be estimated from the data using assumptions for shape and density, they do not differentiate the fraction of DPM that is EC.

There are a variety of instruments available to estimate the carbon concentration in aerosols. These include the Magee Scientific Aethalometer, the Thermo Electron MAAP, the Photoelectric Aerosol Sensor (PAS 2000) from EcoChem Analytics, and the Series 5400 Ambient Carbon Particulate Monitor from Rupprecht & Patashnick (*8*). The Aethalometer has been shown to be effective in estimating the EC in DPM. It determines carbon mass concentration in sampled aerosols by measuring the optical light absorption of black carbon (BC) at specified wavelengths (*9*) and gives results that correlate very well with EC concentrations evaluated by the NIOSH Method 5040 (*10*).

Given the need to measure EC generated by diesel engines in underground maintenance shops, rather than using some combination of the instruments mentioned above, we chose to evaluate direct-reading photometers. Photometers are simple and inexpensive devices that give readings that have been shown to correlate well with total particulate mass concentration measurements. The main aim of our work was to evaluate the effectiveness and accuracy of the photometers for measuring EC at the engine tailpipe. This was done by taking measurements of undiluted diesel exhaust using three identical direct-reading photometers while simultaneously collecting three filter samples of the exhaust for analysis using the NIOSH Method 5040. Using the NIOSH method as a reference, the data were subsequently analyzed to determine how well the photometer readings approximated the EC measurements. This approach is similar to a study conducted in Australia, testing multiple instruments against filter samples, including the instrument used in this study (*11*). That study was conducted on diluted exhaust, however, while our work was designed as a preliminary feasibility study regarding the use of photometers to monitor EC emissions directly at the tailpipe, i.e., using undiluted exhaust. Based on the preliminary results presented here, we are planning further testing of our method on a variety of different engines in the future.

2. Materials and Methods

2.1. Direct-Reading Photometers. Portable, direct-reading dust monitors are often called nephelometers, aerosol photometers, or light scattering aerosol monitors. They operate by illuminating aerosol passing through a defined volume and detecting the scattered light (*12*). Such devices have been shown to yield measurements which correlate well to gravimetrically measured mass concentrations of airborne particles (*13*, *14*). However, when measuring a polydisperse aerosol containing ultrafine particles not detectable by photometric techniques, a calibration factor may be used to account for the undetectable portion. Best data correlations are achieved when such calibration factors

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account for variations in particle size and refractive index, which is influenced by chemical composition. It is therefore recommended that such devices be calibrated against a standard measurement method for the aerosol of interest under the same test conditions (*12*).

There are many different photometers, each with their own unique operating parameters and calibration scheme (*12*, *13*). The photometer chosen for this evaluation is the Thermo Electron Personal DataRAM 1200 (PDR). We chose the PDR because of its relatively low cost and ease of use. The PDR is a small lightweight instrument designed to be attached to a workers belt and is often operated as a passive sampler. It can also be used as an active sampler as was done for our experiments. For active sampling, an auxiliary pump was used to draw air over the sensor and a cyclone (BGI model GK2.05) was attached to remove larger particles, i.e., those larger than 5.5 μm. The PDR gives best sensitivity in the respirable range, is auto ranging between 0.001 and 400 mg/m^3, can sample as fast as 1 Hz, and has built-in data logging capability. It is rezeroed by simply resetting the zero reference and can operate approximately 70 h at an average aerosol concentration of 5.5 mg/m^3 before the zero reference begins to drift due to contaminated optics (*13*). Rezeroing is obtained by running a clean flow of air through the PDR using a high efficiency particulate (HEPA) filter, while the PDR recalibrates itself by resetting the zero reference. It also is field-cleaned by clearing the optics with a compressed air jet. The PDR is factory calibrated using SAE Fine (ISO Fine) dust, provided by Powder Technology Inc. which conforms to ISO 12103 Pt1 (*15*).

2.2. NIOSH Standard Method 5040. The mass concentration of the EC and OC was determined using the NIOSH Method 5040, although only the EC data are reported here. The NIOSH Method 5040 is often used when assessing the EC content of diesel-laden aerosols and is considered a reference method (*16*). In this method, the particulate is collected on quartz-fiber filter media and analyzed using a thermal-optical transmittance technique (*17*). This approach is somewhat similar to the interagency monitoring of protected visual environments (IMPROVE) thermal optical reflectance method but with different temperature and charring correction protocols (*18*). Note that the NIOSH method is a thermal−optical transmittance method (i.e., it uses the optical transmittance to correct the charring), while the similar IMPROVE method is called a thermal−optical reflectance method and uses reflectance of the optical signal to correct for the charring (*19*).

For analysis by the NIOSH method, samples were collected at a flow rate of 1.94 Lpm onto prefired quartz-fiber filters mounted in 37 mm plastic filter cassettes. Stable flow was achieved by drawing exhaust emissions through critical orifices using a vacuum pump. For each test condition, filter samples were collected in triplicate to ensure repeatability of results, and sampling times were 20 min in length to ensure adequate filter loading. The filter loadings for all tests ranged from 27 to 180 μg/cm^2 for EC. The EC data presented in this report are based on the average of the three samples taken at each condition. The samples were sent to a NIOSH-contracted lab (DataChem Labs in Salt Lake City), along with media blanks and field blanks as required. The samples were analyzed by the thermal optical method (*20*) and time-weighted average values of EC and OC in units of mg/m^3 were derived from the analytical results.

The NIOSH Method 5040 utilizes a thermal−optical technique to quantify the EC and OC. During implementation of the method, carbon volatilized below 850 °C in a helium atmosphere is defined as OC, while carbon burned off at 850 °C in an atmosphere of oxygen and helium is defined as EC. The optical component of the method entails evaluation of the char, i.e., residue left on the filter due to pyrolysis of some OC during the first heating cycle. This char is oxidized at the beginning of the second heating cycle and its mass apportioned as OC. An optical technique is used to determine when this char is removed from the filter, which is why the method is called “thermal−optical”.

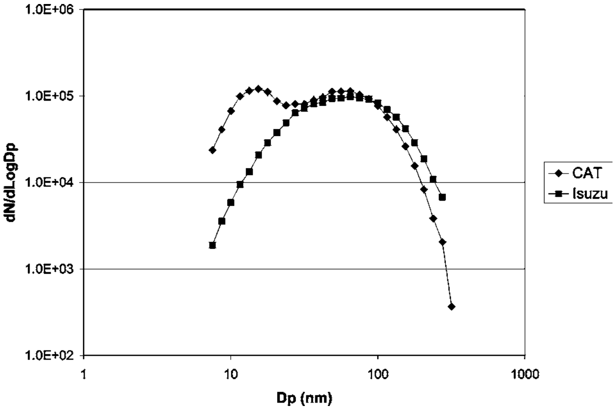

FIGURE 1. Number-weighted particle size distribution (dilution-corrected) for Isuzu engine and Caterpillar C-12 engine, both measured with an SMPS at moderate loads.

2.3. Diesel Particulate Generation. The source of diesel particulate for this work was an Onan-Cummins “Quiet-Diesel” genset powered by a three cylinder, 1.5 liter Isuzu engine. This unit is capable of providing 10 kW of continuous AC power at a fuel flow rate of about 4 kg/hr. To maintain a steady 60 Hz of AC current from the generator, the fuel flow to the engine is controlled by an electronic governor actuator that maintains a constant engine speed of 1800 rpm. The load on the engine is provided by loading the generator outlet with a resistive load bank made by the Simplex Company. For the experiments, the engine was run at three conditions: no-load (0 kW), 2 kW, and 6 kW. The corresponding fuel flow rates were 1.0, 1.4, and 2.3 kg/hr, respectively. The fuel used for all tests was standard no. 2 diesel fuel with nominal sulfur level of 350 ppm. It should be noted that the size distribution of particles generated by this relatively small engine (Figure 1) varies somewhat from that of typical heavy duty engines, i.e., under normal operating conditions it produces no discernible nuclei mode. This does not obviate the application of results to larger engines, however, since the photometer is insensitive to the ultrafine particles.

The emissions sample is drawn from the tailpipe as shown in Figure 2. The 3.8 cm diameter exhaust line is connected to a 20 cm diameter vertical vent pipe. About 15 cm from the muffler exit, a sample is drawn through a tee fitting in the exhaust line at a flow rate of about 20 Lpm. The sample is routed through a 2.4 m long, 1.3 cm diameter insulated pipe, then through a 6.1 m long, 1.3 cm diameter copper cooling coil to the multiport sampler.

2.4. Sampling Logistics. The primary goal of this work is to evaluate the PDR photometer in parallel with samples taken using a standard method to directly evaluate the accuracy of the instrument. To that end, simultaneous measurements were made at the diesel tailpipe using both approaches with the engine operating at three different loads: 0, 2, and 6 kW.

During each test, three samples of EC for analysis were collected simultaneously onto prefired quartz filters mounted in cassettes, using calibrated critical orifices to control flow rate, set to 1.94 Lpm. Two types of such filter samples were collected at each engine condition, each using a different type of filter holder. One was a DPM cassette no. 225−317 with a submicron impactor made by SKC Inc., and the other was a standard 37 mm filter cassette. It was found that the difference between the two approaches was negligible, so

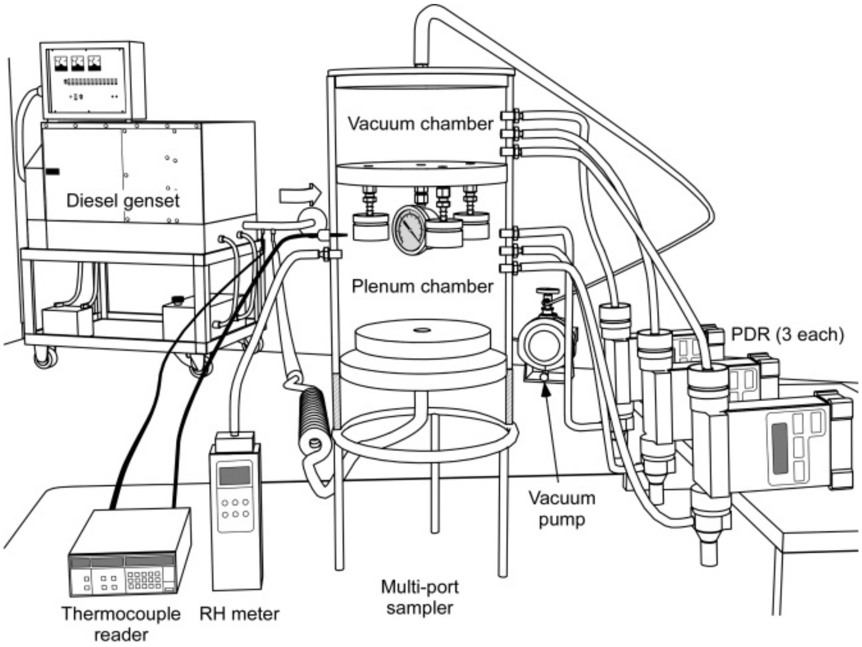

FIGURE 2. Experimental setup.

only the data from the standard 37 mm cassettes are presented in the results section.

For all tests, three PDRs connected in parallel sampled the exhaust emissions through calibrated critical orifices at the same flow rate (1.94 Lpm) as for the three filter samples. The orifices were located on the exhaust side of the PDRs to allow the instruments to operate at atmospheric conditions. Connected to the upstream side of the PDRs were 2.5 μm BGI Model GK2.05 cyclones which provided a convenient connection to the PDR, as well as ensuring that no large particles entered the detection region. Since very little of the DPM is greater than 2.5 μm it is likely that the system would have worked without the cyclone in place. For the tests where the SKC filters were used, the PDRs were also equipped with their own custom submicron impactor to provide similar sample-size selection. This customized impactor was made by dismantling an SKC filter cassette, removing the filter and leaving the impactor, and reconnecting it to the PDR after reassembly. Impactor seal integrity is assured with an O-ring.

For collecting parallel samples on multiple filters as well as with the PDRs, we built a multiport sampler. It consisted of a cylinder with an inlet and multiple outlet holes (Figure 2). The cylinder was divided into two sections. The larger inlet section provided a mixing baffle and a plenum chamber, while the smaller top section provided the vacuum. A thick plate separated the sections and was fitted with critical orifices that act as flow meters for the filter samples. The filters, PDRs, and other instruments were connected to the multiport sampler so that they measured the same aerosol at the same time. A vacuum pump was used to maintain the vacuum for the critical orifices, and a vacuum gauge monitored the pressure to ensure that the orifices reached critical flow. The temperature and relative humidity in the multiport sampler were monitored, as well as the exhaust gas temperature, which was measured by a thermocouple inserted into the tailpipe.

The exhaust emissions underwent minimal treatment before being sampled. Since our goal was simplicity, we sampled the undiluted exhaust and only cooled and dried it with a simple condenser, which consisted of a coil of copper tubing. Water collected in the coil and was emptied manually between tests to prevent water build up in the coil, but a water trap could easily be installed in such a setup.

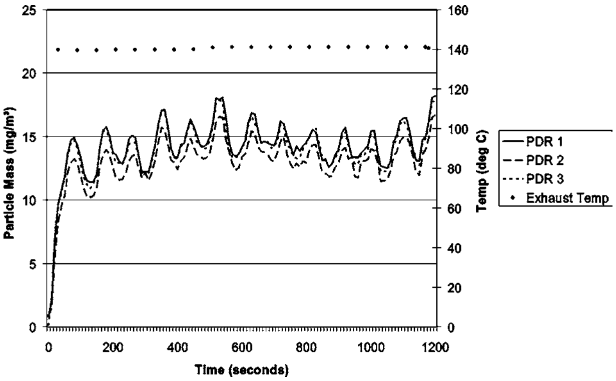

FIGURE 3. Raw PDR data from a single test (0 kW engine load).

3. Results and Discussion

3.1. Raw PDR Data. Figure 3 shows an example of real-time PDR data collected during one of the 0 kW experiments which is typical of the type of data collected at all loads during these tests with the PDRs. Note that all three PDR's are fairly consistent (less than 15% difference between instruments) and that the signal is somewhat sinusoidal. This trend appeared in other instruments we used during the course of this work, including the TSI DustTrak photometer and the EcoChem particle-bound polycyclic aromatic hydrocarbon (PAH) monitor, suggesting it is a real phenomenon. The sinusoidal readings are possibly related to the variation in soot production as a function of fluctuations in engine block temperature via the cycling of the thermostatically controlled cooling system. This highlights an advantage to using the PDR for collecting such data under field conditions, since it is easy to see fluctuations in the data which might indicate unwanted changes in the test conditions.

3.2. Time-Weighted Average Data. For each test, this real-time data from each of the three PDR's was averaged over the duration of the test to get a time-weighted average for each PDR. The average of those three numbers yielded a single mass concentration value for each test. Likewise, the data from the three filter samples collected for each test was averaged. An example of such averaged, uncorrected data is shown in Figure 4. Note that the data from the triplicate samples show relatively little variation. The relative standard

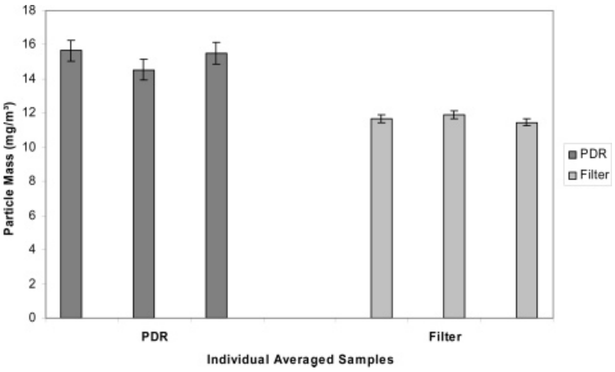

FIGURE 4. Averaged EC filter data over a 20 min test and PDR data for the 2 kW engine load with standard deviation bars shown.

deviation for the three averaged PDR measurements was approximately 4% and for the three filter measurements approximately 2%.

Time-weighted averages of raw data from four different tests for each load are plotted in Figure 5 and demonstrate that although there is variation in the data, some trends are clear. First note that since the NIOSH 5040 method is considered a reference method, the filter data is assumed to reflect the actual EC concentration in the exhaust aerosol. The fact that the PDR data varies from the actual EC levels is due to errors brought on mainly from two sources: instrument calibration and relative humidity (RH) effects. Also note that the test-to-test variation is highest for the low load. This is believed to be a function of how long the engine had been running, which affected engine block temperature that was not monitored. Some tests were done as soon as the tailpipe temperature stabilized after start up, whereas others were done after the engine had been running at higher load. At low load, the carbon emissions are highest which verifies earlier work using this engine (*21*). At high load, the emissions are reduced, especially the measured EC. For this case, the fuel consumption is greatest, and the measured relative humidity of the emissions is thus highest, which helps explain the higher PDR readings as the load increases.

3.3. Correlating the Data. The raw data from the photometers were next plotted against the data from the NIOSH method 5040 (Figure 6) and show that the correlation has a linear trend for each load condition, but that the slope varies. This is expected since both the size distribution and the EC/OC ratio change as the engine load increases, affecting the scattering efficiency and the refractive index, respectively. This confirms the importance of calibrating photometers to the aerosol of choice as demonstrated in earlier works using various types of dust (*13*). These correlations are also affected by RH, which requires correcting the data using RH measurements made at each engine load condition.

TABLE 1. Relative Humidity and its Corresponding Correction Factors

relative humidity (%)	0	40	50	60	70	80	90	100
correction factor	1.0	1.0	0.93	0.83	0.71	0.56	0.40	0.22

Correction of the raw PDR data entails two steps. First, the data must be corrected for RH effects, since the PDR output is affected when RH > 50% (*22*). Second, the humidity-corrected data must be compared to a standard (in this case, the filter data) and a calibration factor calculated to correct the data to the standard. The PDR is factory-calibrated using a reference dust (*15*), but also allows for user-based field calibration. The calibration consists of entering a calibration factor (via the menu-driven graphical user interface), which is a multiplier used to correct the signal intensity and thereby adjust the instrument output. In this case, the calibration factor accounts for the differences between the diesel aerosol and the aerosol used for factory calibration.

We first determined an appropriate multiplier to correct for the RH levels measured at each engine load, using the data in Table 1, published previously (*22*). Note that for each set of tests at a specific load condition, RH levels were approximately the same: ~50% RH at 0 kW, ~70% RH at 2 kW, and ~90% RH at 6 kW.

We then calculated the ratio of the EC values (the standard) and the RH-corrected PDR data for all tests and averaged them to get another such multiplier (i.e., a single calibration factor for the exhaust aerosol from this engine). Using both multipliers to adjust the PDR data gives the best possible estimate of EC. A linear regression of the corrected PDR data versus the standard method filter data is shown in Figure 7. Note that the data correlate well, as the correlation coefficient (R^2) is near unity (0.97) and the regression slope (y) is slightly below unity (i.e., 0.89).

The same process was repeated for estimating the correlation between the total carbon (TC) (where TC = EC + OC) and the PDR data, yielding a correlation coefficient of 0.96 and a regression slope of 1.12 after correcting for RH. The variation in the regression slopes derives from differences in the calibration factors for EC versus TC. This data is not shown since our focus was on the EC fraction. The OC data may have been slightly artificially high since we did not use

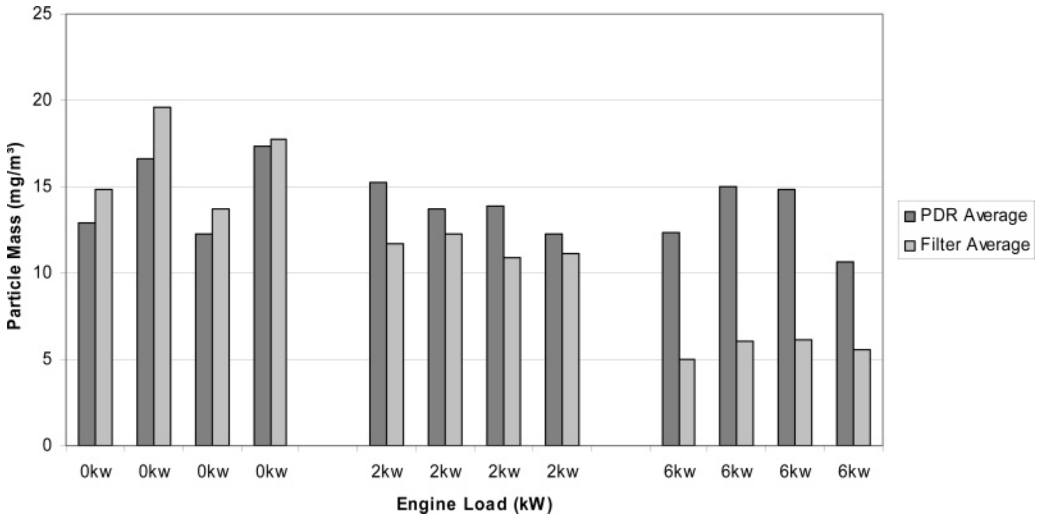

FIGURE 5. Averaged uncorrected PDR and EC filter data from multiple tests.

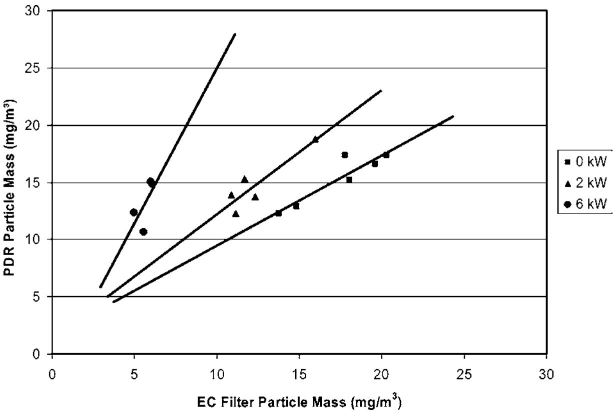

FIGURE 6. Averaged uncorrected PDR data versus standard method (EC filter data) for three engine loading conditions.

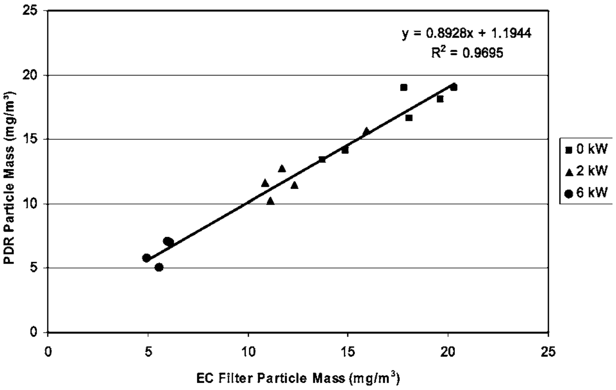

FIGURE 7. Linear regression of corrected PDR data and EC filter sample data.

a “dynamic blank” to correct for potential condensation of OC vapors on the filters during the collection process. The difference in regression slope may also suggest that the calibration factors reflect higher levels of OC on the smaller particles as demonstrated in previous studies (*23*). More research is recommended to better understand this phenomenon.

It is important to note the limitations of this method. The PDR will measure more than just the elemental carbon particulates and anything affecting the relationship between the elemental carbon and other particulates could affect the calibration factor. Also the calibration factor will vary for each engine and through use of different fuels or control technologies such as diesel particulate filters. This outlines the importance of verifying the results using a reference method to ensure the best accuracy.

Results of this preliminary study show that, under laboratory conditions, measurements made with the PDR photometer can be used to estimate EC emissions reasonably well if the PDR data is properly adjusted for RH effects and calibrated to a standard. Such measurements can be done with no dilution of the exhaust emissions and by using a simple condenser to cool and dry the sampled exhaust enroute to the instrument. Photometers such as the PDR may thus be a useful tool to evaluate trends in engine emissions (particularly EC) over time and for comparing relative levels of EC generated by different vehicles in a fleet. More testing is necessary to evaluate the magnitude of day-to-day variations and the practicality of sampling raw exhaust under field conditions to determine the applicability of this method for use in emissions-assisted maintenance programs involving a variety of engine and vehicle types.

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