Novel Approaches to Measure Diesel Emissions

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Abstract

Due to its excellent reliability and fuel economy, diesel-powered equipment is used extensively in metal and nonmetal mines throughout the world. Most of the research to date suggests that the majority of fine particulate matter (FPM) in underground mines is derived from diesel exhaust. The US Mine Safety and Health Administration (MSHA) has been enforcing an FPM concentration limit of 400 $\mu g/m^3$ total carbon (TC) (or 308 $\mu g/m^3$ elemental carbon [EC]) in mine air and is considering lowing it to 160 $\mu g/m^3$ TC by January 2006. Diesel exhaust also contains gaseous hazard pollutants, such as carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2), and volatile organic carbon (VOC). The emission rates of these pollutants depend strongly on the fuel, load, type, and age of the diesel engine. Concerned with the mining operation and miner's safety, both time-integrated and peak emissions from mining diesel equipment need to be accurately quantified.

Emission measurements with conventional dynamometer setups have the advantage of testing engines under a variety of running cycles. However, only a very limited number of vehicles can be tested, and often they do not represent the entire fleet (Moosmüller et al., 2001a; Moosmüller et al., 2001b). The Desert Research Institute (DRI) has developed in-plume and remote sensing measurement methods that allow different pollutant concentrations in the exhaust to be measured by normalizing their concentration to the excess carbon dioxide (CO_2) concentration measured. Knowing the carbon content of the fuel combusted, this ratio can be related to the amount of fuel consumed (Moosmüller et al., 2003), and fuel-based emission factors can be derived. The DRI FPM remote sensing system uses a 266-nm ultraviolet laser light to detect the backscatter signal and total path extinction across the plume. With suitable assumptions regarding size distribution and particle composition, particle mass emissions are calculated for FPM. Concurrent measurements of CO, HC, and NO_x, emissions are made with a commercial remote sensing device. This method allows the measurement of fuel-based emission factors for many vehicles driven through a checkpoint. The vehicle running conditions are usually consistent at the check point (e.g., cold start, hot stabilized, etc.). For in-plume measurements, vehicle exhaust is drawn into a manifold where the sample stream is directed to an extractive Fourier Transform Infrared Spectrometer (FTIR) to measure chemical composition as well as to several particle measuring instruments including nephelometers to measure light scattering, a photoacoustic instrument to measure light absorption, particle counters to measure particle size distributions, and filter samplers to measure PM_{10} and $PM_{2.5}$ mass concentration and chemistry. The FTIR quantifies multiple species including CO_2, CO, NO, SO_2, NH_3, and HC in the plume. EC concentration can be directly derived from the photoacoustic measurement. The in-plume method can examine many vehicles in a short time as well, and allows more species to be determined.

These monitors have been successfully applied to quantify emissions from both on-road and off-road vehicles. For on-road mobiles sources, the test stand is deployed on the side of the road to sample a large number of vehicles passing the monitor. Emissions from off-road vehicles or engines are measured by installing the test stand inside a cargo van with a sampling inlet positioned on top of the vehicle. The applicability of these methods to study mining diesel emissions will be discussed.

Keywords: mining diesel, elemental carbon, remote sensing, photoacoustic

1. BACKGROUND

Diesel-powered equipment is used extensively in metal and nonmetal mines throughout the world. Although there is debate on the exact contribution of diesel emissions to airborne fine particulate material (FPM) (particle material <2.5 μm aerodynamic diameter), in underground mines, most of the research to date suggests that the majority of FPM is derived from diesel exhaust, with the remainder composed of re-suspended dust, drilling oil mists, and cigarette smoke (Haney 1992; Cantrell and Watts 1997). Surveys of mine air have shown that fine particle mass in underground mines can exceed 1 mg/m^3 of air (Cantrell, Rubow et al. 1993; Tomb and Haney 1995). This is significant because diesel exhaust has received much attention regarding its potential effect on human health in ambient and occupational exposure conditions (NIOSH 1988; IARC 1989; Cal/EPA 1998).

Diesel exhaust emissions factors are usually derived by sampling exhaust from an engine operated outside of its vehicle on an engine dynamometer. Various levels of power and water-braking are applied to simulate how the engine would respond in actual operations. One of these cycles, the Federal Test Procedure (FTP, U.S. EPA, 1978), is used to ascertain that engine/fuel combinations meet EPA emissions standards (U.S. EPA, 1997a). Engine dynamometer test cycles are used to estimate emissions factors in units of g/bhp-hr (1 horsepower=0.7457 kilowatts), which are usually translated into vehicle miles traveled (VMT) for on-road vehicles. PM, NOx, CO, and HC emission rates are measured by integrative methods over certain parts of the test cycle. This methodology has several limitations for on-road diesel engines and is inadequate for non-road engine uses. In the Air and Waste Management Association's 2001 Critical Review (Lloyd and Cackette, 2001) and Discussion (Chow, 2001), Dr. Alan Lloyd, Chairman of the California Air Resources Board (ARB), noted the poor status of on-road diesel emissions estimates, stating that California's entire diesel inventory is based on fewer than 50 engine dynamometer tests acquired over the past two decades. Even worse, they observed that emissions estimates for off-road diesel engines were typically extrapolated from on-road engines, fuels, and cycles and that better "real-world" emissions measurements were needed for non-road applications, such as the use of diesel engines in the mining industry.

There are other issues related to the diesel particulate matter (DPM) measurement. On January 19, 2001, the U.S. Department of Labor's Mine Safety and Health Administration (MSHA 2001) initiated new rules to establish health standards for underground metal and nonmetal mines in which diesel equipment is used. The rules establish a concentration limit to be phased in over a 5-year period. Under these rules, an interim limit of 400 μg/m^3 of total carbon (TC) went into effect July 19, 2002. The exact effective date for a final limit of 160 μg/m^3 is currently under review. This standard based on TC is aimed at measuring and regulating the contribution of DPM to mine air. On June 6, 2005, MSHA published a final rule

(MSHA 2005), which converted the interim concentration limit to 308 μg/m^3 of elemental carbon (EC). This critical change from TC-based to EC-based regulation reflects the fact that EC is a better marker for diesel emission than TC. In metal or non-metal mines, drill oil mist, environmental tobacco smoke (ETS), and organic solvents also contributes to TC in the mine air, mostly in the form of organic carbon (OC). MSHA (2005) states "no reasonable method of sampling that could eliminate the interferences from oil mist or that would effectively measure DPM levels in the presence of ETS with TC as a surrogate". While EC is a component of TC (a dominant component in DPM), the accuracy for quantifying the TC:EC ratio is not yet satisfactory. Conventional thermal/optical methods do not give consistent results.

In this paper, we present innovative in-plume and cross-plume systems for quantifying diesel exhaust emissions. Both of these systems differ from conventional dynamometer tests by characterizing emissions in real time and determining emission factors within a very short test duration. Since usually no owner consent is required, many more vehicles can be tested to determine average emissions for a registered fleet. The emission factors are fuel-based, and therefore the total emissions (inventories) can be calculated directly from the fuel consumption. These systems are designed to quantify multiple species, including all major gaseous and particulate pollutants in the diesel exhaust. The in-plume system contains a photoacoustic sensor that can detect EC in real time without being interfered by OC (TC = OC + EC). These systems are very suitable for measuring mining diesel emissions.

2. METHOD DESCRIPTIONS

2.1 Cross-plume remote sensing system

The particulate matter (PM) vehicle emission remote sensing system (VERSS) was developed at DRI for remote sensing of particle emissions from vehicles (Moosmüller et al., 2003). It uses 266-nm ultraviolet laser light for greater sensitivity to scattering from sub-micrometer particles. An optical pulse leaves the laser source, is partially backscattered by particles in the exhaust plume, and is then detected by a sensitive photo multiplier tube collocated with the laser source. The advantage of using a Lidar system is that the reflected signal can be associated with particles at a specific range *r*. With suitable assumptions regarding size distribution and particle composition, the Lidar backscatter signal and total path extinction are used to estimate particle mass emissions.

Figure 1 shows a schematic diagram of the PM VERSS system. PM VERSS is triggered by a beam unblock signal that follows a beam block signal of 0.2-0.4 second, indicative of a passing car. The transmitter contains a Nd:YAG laser (λ=1.064 μm), frequency-quadrupled to yield 266-nm ultraviolet light. A beam-expander is employed to sample a larger cross section of the plume; this also mitigates eye and body safety concerns from the laser. Laser pulse duration is approximately 1 ns, providing 15-cm wide imaging distances across the plume. The receiver employs a Newtonian telescope to collect the backscattered light. Light is passed from the telescope to a photo-multiplier (PM) tube. A solar-blind filter eliminates background light. A fast response pre-amplifier increases the signal amplitude for input to a data acquisition system. The data acquisition system uses a 1 GHz, 2 Giga-sample-per-second oscilloscope to capture the backscattered UV light. Computer processing includes background subtraction, separating tire road dust backscatter from exhaust backscatter, and linking the PM VERSS and RSD-3000 measurements from the same vehicle.

The Remote Sensing Device (RSD) 3000 is a commercially available unit for measuring mobile emissions of CO, HC, and NO_x. The system generates and monitors a collinear beam of infrared (IR) and ultraviolet (UV) light emitted and reflected approximately 30 cm and 50 cm above a single lane road. Vehicles drive through the beam and the exhaust causes interferences in the amount of light transmitted to the detector. These interferences are related to the concentrations of pollutants in the vehicle's exhaust. The system is equipped with a speed and acceleration system to measure the specific power of the vehicle as its exhaust is being monitored (Jiménez et al., 1999). A digital image of the vehicles' license plate can be acquired with each measurement so that emissions can be related to vehicle information such as fuel type and model year. DRI's RSD3000 has been modified from it original configuration to accommodate the measurement of emissions from elevated vehicle exhaust stacks. The system can be mounted on an elevated platform so that the exhaust plume is emitted directly into the optical path. Although the vehicle itself never blocks the exhaust sampling beam, a separate trigger is used to initiate sampling as the vehicle passes the test section. Remote sensing of CO, HC, and NO is a proven technology and has been used to measure emissions from both gasoline and diesel powered vehicles for over a decade (Stedman, 1989).

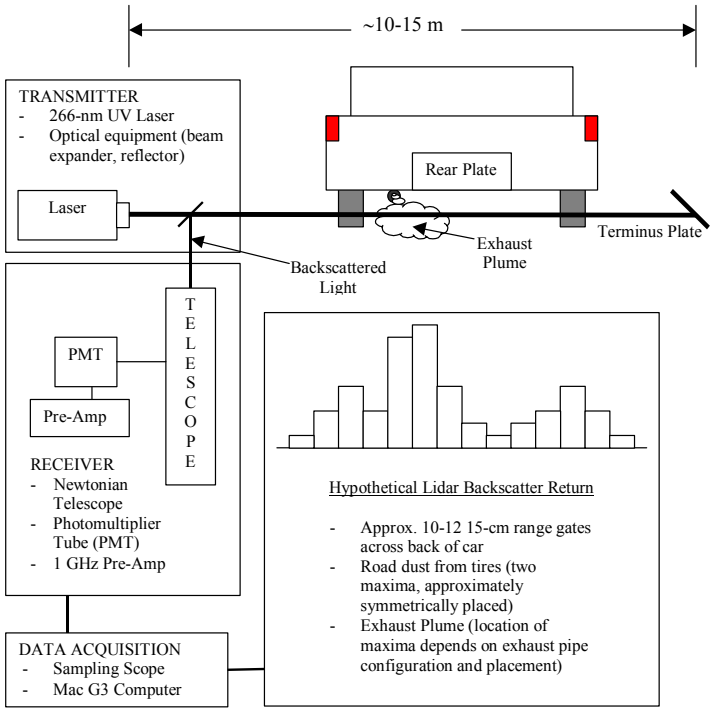

Figure 1. Schematic diagram of the VERSS particle detection system consisting of: 1) the transmitter, a UV laser; 2) the receiver, which includes a Newtonian telescope, a photo-multiplier tube, and a 1 GHz pre-amplifier; and 3) the data acquisition subsystem, which includes a 1 GHz, 2 Giga-sample oscilloscope and a computer.

The system is being used to make roadside exhaust particulate measurements for emissions from moving vehicles, both spark ignition and diesel. Average backscatter signal and CO_2 concentration for a single plume are ratioed and related to fuel-based PM emission factors by normalization to collocated in-plume samples for a subset of engines, fuels, and operating conditions. Figure 2 shows emissions ratios for four vehicles using the PM VERSS and the RSD-3000 remote sensors. Notice that there is a substantial difference in particle emissions from these vehicles, with the Lincoln Continental having the highest emissions per unit of fuel

consumed (as indicated by the CO_2 emissions). Particle emissions are not well correlated with gas emissions. The highest CO emissions that are usually the main focus of inspection and maintenance programs, do not necessarily correspond with high PM emissions.

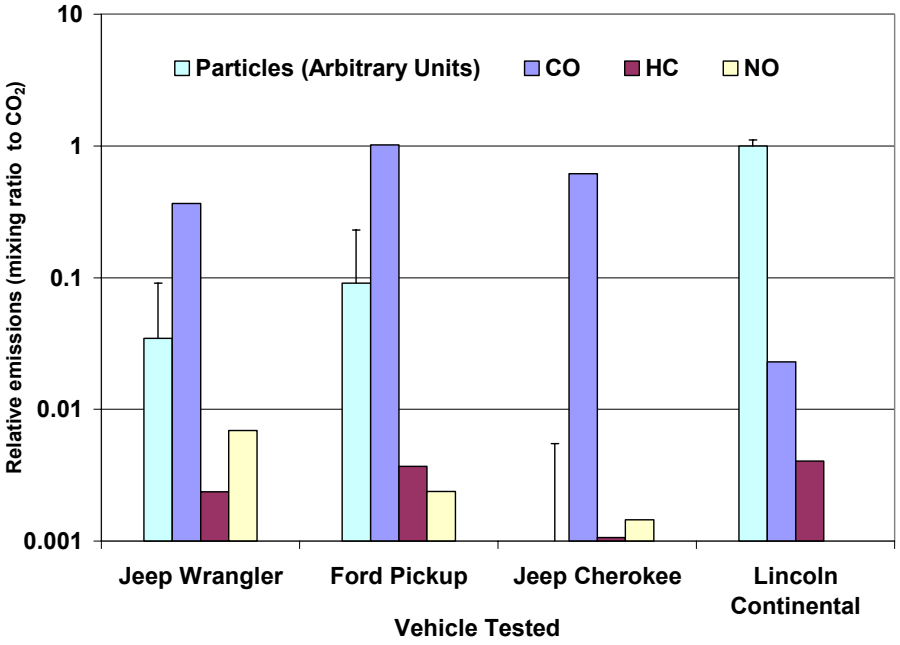

Figure 2. Example of particle mass, carbon monoxide (CO), hydrocarbon (HC), and oxide of nitrogen (NO) ratios to carbon dioxide (CO_2) measured with the prototype VERSS particle sensor and the commercial RSD-3000 gas sensor. The bar represents the uncertainty of the particle emissions, as indicated by the standard deviation of repeated measurements in the same exhaust plume.

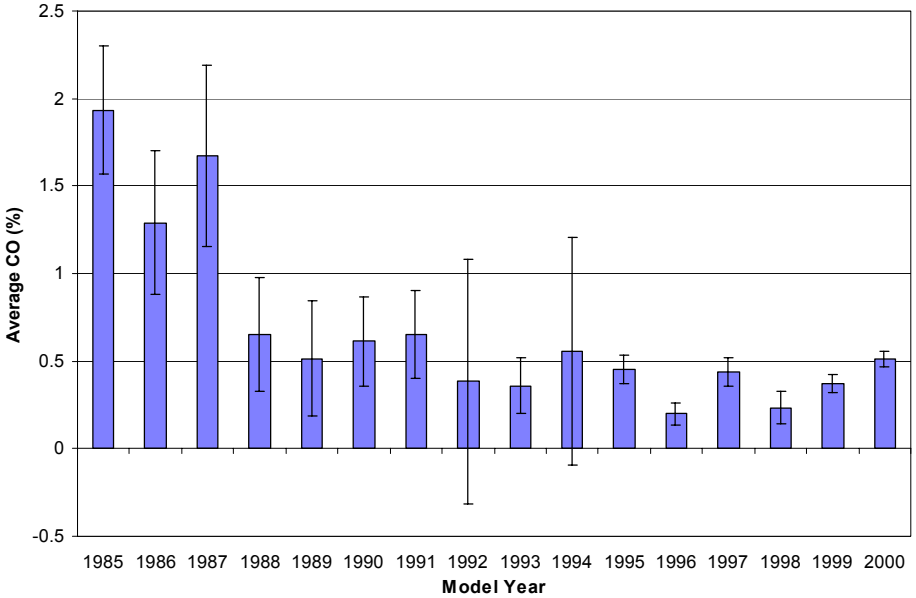

Figure 3. Example of how average emission factors can be stratified by model year for carbon monoxide emissions. These on-road emissions allow decision-makers to target emissions reduction strategies at those vehicles and operating modes that make the largest contributions to overall emissions.

For on road vehicles, particle remote sensing shows that 80% of the vehicles are relatively low emitters and that most of the emissions come from a few vehicles, either because they are in a high emissions operating modes or because they are persistently high emitters. This is also likely the case for mining vehicles and equipments. Figure 3 shows how the data acquired by on-road testing in many different situations can be stratified. In this case, the stratification is by model year. Average emissions factors, similar to those in Figure 3, as well as emissions distributions (for different percentiles) can be included in the emissions estimation software to permit "what-if" questions to be answered when evaluating control strategies. For example, a software tool allows determining the effect on overall emissions of replacing all pre-1990 engines with newer engines, or of identifying and repairing/removing units that are the highest 10% of emitters.

Both the RSD3000 and the PM VERSS can be used to sample emissions from mining vehicles capable of being driven through an emission test section, including drill jumbos and rock bolters. The remote sensing equipment has been operated in two separate configurations to sample exhaust from tail pipes and elevated exhaust stacks. To obtain the emission characteristics as a function of driving cycles, vehicles can be driven repeatedly through the remote sensors for a variety of controlled conditions (i.e., cold start, coasting/idling, constant speed, and heavy loading). Additional tests need be conducted to measure emissions from undrivable diesel equipments.

2.2 In-plume system

The in-plume sampling system consists of an emissions monitor test stand that employs multiple gas and particle sensors for CO_2, CO, NO, SO_2, NH_3, PM_{10}, $PM_{2.5}$, BC, and HC in the plume of mobile sources (Nussbaum et al., 2005). An extractive Fourier Transform Infrared Spectrometer (FTIR) equipped with a liquid nitrogen cooled Mercury-Cadmium-Tellurium (MCT) detector is capable of measuring gaseous concentrations at the 500 ppb level at one second intervals. Figure 4 shows a time series of the CO_2 and NO emissions from this test vehicle passing the instrument. A gasoline powered 1979 Chevy Van was driven at approximately 20 km/hr past an FTIR spectrometer outfitted with a 20-m optical path gas cell. The vehicles exhaust pipe was located 5 m upwind of the FTIR instrument. The ratio of measured pollutant concentrations to CO_2 is proportional to the fuel consumption emissions factor. In Figure 4 the regression of NO versus CO_2 indicates a correlation coefficient of 0.98 and slope of 0.019 ppm/ppm.

For real world measurements, engine exhaust is drawn into a manifold where the sample stream is directed to the FTIR as well as to several particle measuring instruments including nephelometers to measure particle light scattering, a photo-acoustic aerosol absorption monitor to measure particle light absorption (elemental carbon), optical particle counters to measure particle size distributions from 0.3 to 20 μm, and an electric low pressure impactor (ELPI) to measure particle size distributions from 30 nm to 10 μm. Filter samplers for measuring PM_{10} and $PM_{2.5}$ mass and chemistry and canister samplers for measuring speciated volatile organic carbon (VOC) from C_2 to C_{12} are used on selected aggregates of engine tests. Figure 5 demonstrates the configuration of the in-plume system.

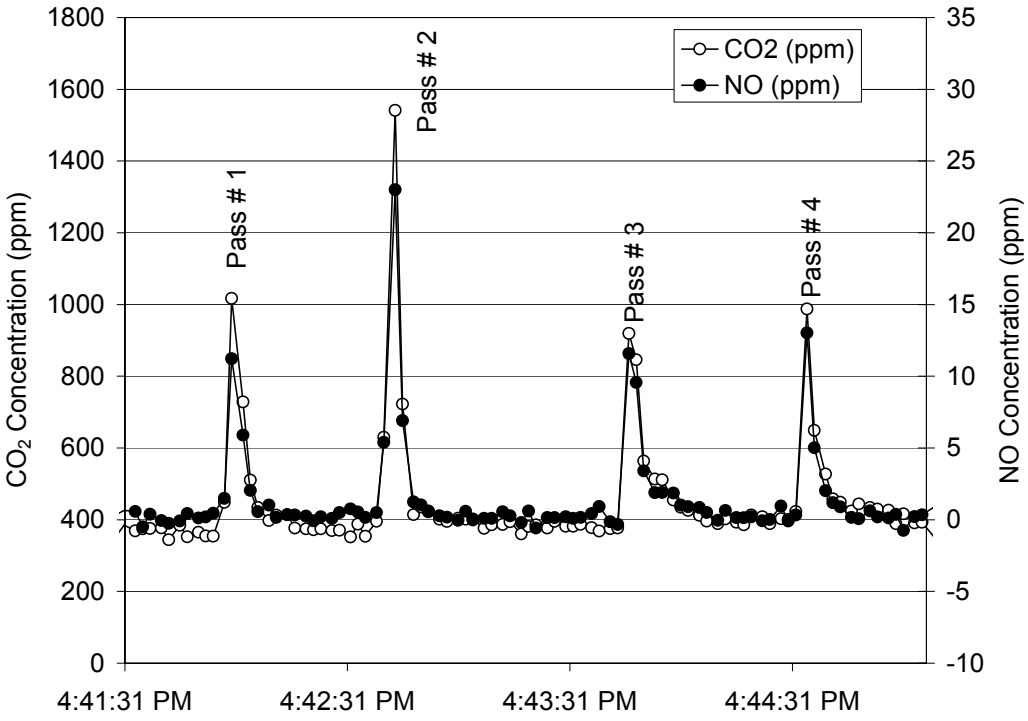

Figure 4. Time series of CO_2 and NO concentrations measured with FTIR downwind of passing vehicle.

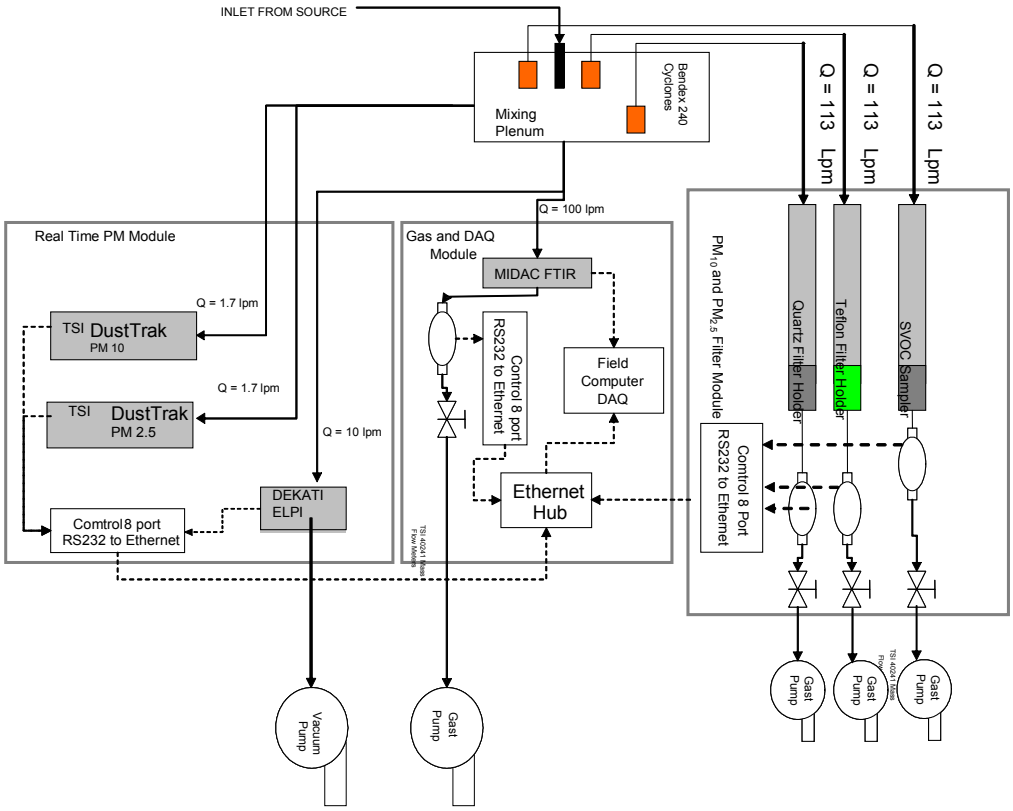

Figure 5. Schematic of DRI In-Plume Sampling System.

For a better characterization of DPM optical properties, a nephelometer and a photoacoustic sensor can be installed in parallel to the ELPI. Nephelometer and photoacoustic sensor detect the light scattering and absorption due to DPM, respectively. The nephelometer, photo-acoustic aerosol absorption monitor, particle counters, and ELPI output data are reported at one-second intervals suitable for correlations with the CO_2 signal from the FTIR. Data from other instruments with slower response times (i.e., filter samplers and canisters) can be associated with the higher time resolution particle measurements to infer fuel-based particle emissions factors for either a large number of vehicles or an extended test cycle from a single vehicle.

To measure gaseous and particulate emissions from mobile and stationary diesel engines, the in-plume monitor can be operated in a variety of sampling configurations. For fixed sources such as power generators, the inlet is positioned in the plume downwind of the source. For on-road mobile sources such as trucks, the stand can be deployed downwind of the road to sample a large number of vehicles passing the monitor. Some off-road vehicles such as tanks and fork lifts operate on less consistent routes than on road vehicles. Emissions from these vehicles can be measured by installing the test stand in the back of a pickup truck with a sampling inlet mounted on a boom in the truck bed. The truck is able to position itself in the plume of the offroad vehicle with minimal impact on the off-road vehicle's normal operation. Under these conditions, emissions factors over a real world test cycle can be directly measured at the source.

2.3 Real-time monitoring of DPM in mines

EC absorbs light in the visible, near infrared, and near ultraviolet regions due to conduction electrons associated with the graphitic structure. EC is therefore often referred to as black carbon (BC). In most situations, EC is the single-dominant light-absorbing component in the atmosphere (Horvath 1993; Watson 2002). OC and dust material may absorb weakly in the visible and near-infrared regions, but their concentrations need to be hundreds times higher than EC to rival EC for light absorption. This fact makes the light absorption coefficient (B_{abs}: the total light absorption cross section per unit volume of air, usually in units of Mm^{-1} where 1 Mm is 1 million meters) a very good indicator for EC, and by association, DPM contributions to particulate matter in the mine air. Since aerosol light absorption by combustion particles occurs throughout the entire particle volume, the light absorption coefficient is proportional to the mass concentration of EC.

The dynamic measurement range must be large for real time instruments that are used to measure aerosol light absorption, and thus EC, in mine conditions. This instrumentation should be insensitive to the interferences from OC, carbonate, and extreme particle loading, as well as to gases present in the mine. It should not in general require a submicron impactor inlet to remove coarse particles prior to analysis. A photoacoustic sensor (Figure 6) measures the light absorption coefficient B_{abs}) in the smoke directly by detecting the acoustic signal pumped by a laser beam through aerosol light absorption (Arnott et al., 1999; Arnott et al., 2000). This instrument can achieve 10-second time resolution with a minimum detection limit (MDL) of 1–2 Mm^{-1}. It should be noted that thermal optical methods (e.g., NIOSH 5040 method [NIOSH, 1999]) require use of a laser to correct for pyrolysis or charring of organic carbon on the filter, and to ultimately determine the split between EC and OC, and the optics of heated particles and filters is nontrivial. There are two additional advantages of using the light-absorption instrument over thermal optical methods: the fine temporal resolution and short analysis/report time. The instrument is designed to measure B_{abs} at a user adjustable time resolution spanning from 1

second to many minutes, and this allows the assessment of not only long-term average DPM concentrations but also short-term peak levels that can be several times higher than the average values. Thermal analysis on filter samples takes weeks to months to accomplish in practice. The light-absorption measurement can be obtained with 1 second time response, which makes it especially valuable for designing and testing mine ventilation strategies for the protection of mining personnel.

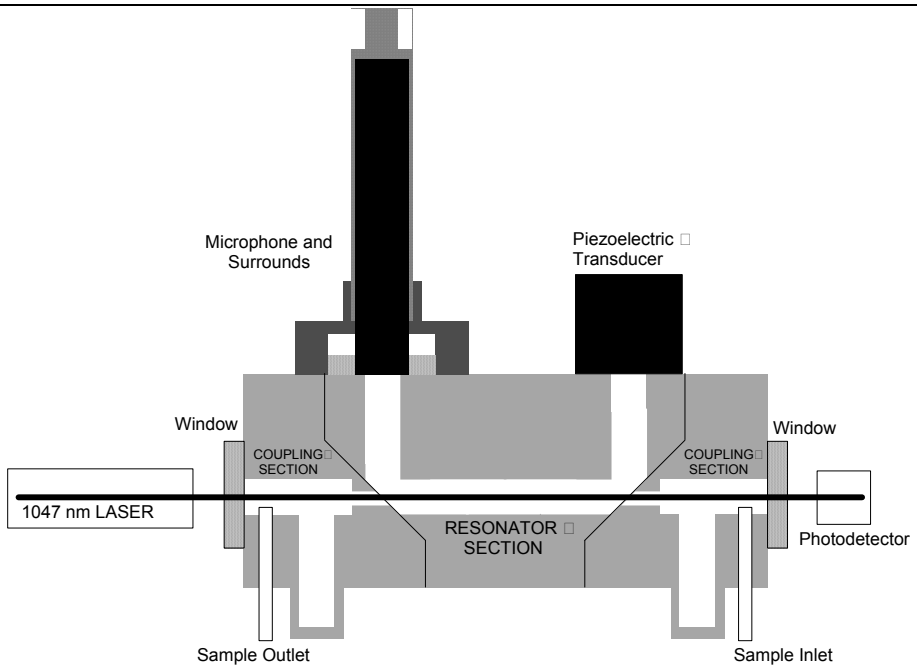

Figure 6. Schematic of the DRI photoacoustic instrument.

An example of the correlation of B_{abs} with quartz-filter-based measurements of EC by the thermal/optical reflectance (TOR) method (Chow, Watson et al. 1993) is shown in Figure 7. Each point is the average EC mass concentration for a particular vehicle driven on a dynamometer under realistic loads. These data were obtained during the National Renewable Energy Laboratory Gasoline and Diesel Split project held in Los Angeles CA during the summer and fall of 2001. This example of the correlation of B_{abs} measured by aerosol light absorption with elemental carbon measured by the TOR technique is typical for diesel samples, and indicates a good comparability between the two methods. Figure 7 shows that all measures of EC determination agree well for the special case of diesel exhaust directly sampled from vehicles through dilution tunnels. EC amount obtained using the NIOSH 5040 method, as programmed into the DRI thermal optical instrument, agrees with IMPROVE method, and the BC measurement obtained with the photoacoustic instrument operated at a wavelength of 1047 nm. In this case the primary exhaust has a large fraction of EC, so that charring and ageing effects are minimized. The agreement of the different methods for EC determination needs to be evaluated for diesel operations in mines where multiple sources of carbonaceous emissions are present.

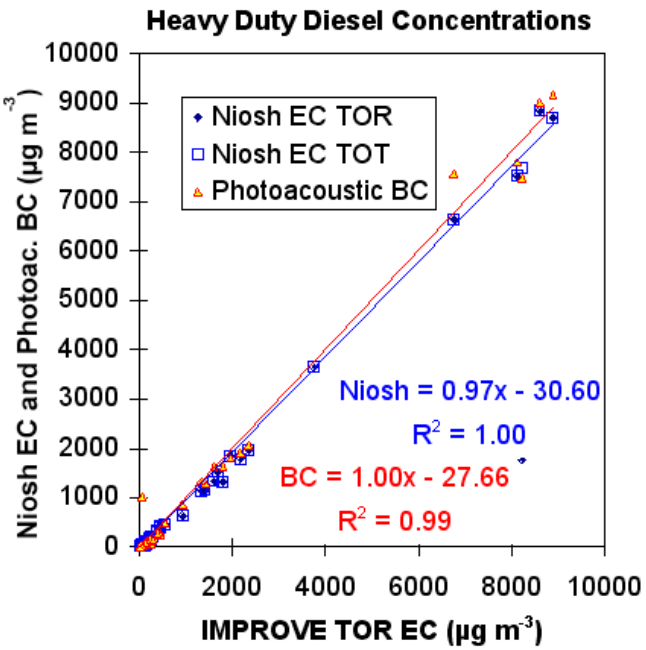

Figure 7. Aerosol light absorption measurements versus elemental carbon mass concentration for heavy duty diesel vehicles tested on a dynamometer. Note the excellent agreement among all methods for the case of directly sampled diesel emissions.

The DPM monitoring instrumentation, including real time (1-5 second time constant) measurement of elemental carbon (black carbon) as well as total particulate mater (TC + Dust), has been developed at DRI and applied to dynamometer testing of gasoline and diesel particulate emissions (Moosmüller, Arnott et al. 2001a; Moosmüller, Arnott et al. 2001b). The speciated PM can be measured in real time include:

- **Elemental Carbon**

— measured by the photoacoustic instrument and Aethalometer behind the nafion dryer inlet, verified by NIOSH 5040 as well as TOR EC.

- **Total PM** = (Elemental Carbon) + (Organic Carbon) + (Dust)

— measured by Dusttrak behind the nafion dryer inlet, verified by TEOM and Teflon filter.

- **Dust**

— measured by the Dusttrak behind the heated inlet. Verified by the Teflon filter and TEOM, when these measurements are performed behind the heated inlet.

- **Organic Carbon** = (Total PM) – (Elemental Carbon) – (Dust)

— measured by Dusttrak – Photoacoustic - Dusttrak on heated inlet, verified by NIOSH 5040 of the quartz filters.

- **Total Carbon** = (Organic Carbon) + (Elemental Carbon)

— Difference between Dusttrak measurements of total PM behind heated inlet. Verified by NIOSH 5040 and TOR of the Quartz filters.

Our near-term goal is to prove the applicability of this sampling suite in monitoring DPM exposure in mines, including both peak and time-integrated levels.

3. CONCLUSIONS

The consistency of DPM measurement is a timely issue as the Mine Safety and Health Administration (MSHA) is replacing the total carbon standard with an elemental carbon standard for quantifying diesel particulate matter for the regulatory interim limit (MSHA 2005: Diesel Particulate Matter Exposure of Underground Metal and Nonmetal Miners. Proposed Rule. 30 CFR Part 57). While the monitoring of mine air quality is important, equal emphasis should be put on measuring emission factors and identifying the major sources of DPM. We have developed innovative in-plume and cross-plume systems to quantify emissions of PM as well as gaseous pollutants from mining diesels. Both of these methods have advantages over conventional dynamometer tests in determining average emissions representative of a vehicle category.

We also propose to base the real-time elemental carbon measurement on an innovative photoacoustic technique, and to evaluate this technique as an alternative to a filter-based measurement method standard. This technique has been shown to be equivalent to the filter-based thermal/optical methods for analyses of primary diesel exhaust dominated by EC. Its performance needs to be evaluated when applied to the challenging conditions in a mine where elevated rock, mineral dust, and oil mists may interfere with the EC measurement. Once validated and calibrated, this real time EC monitor can assess both average and peak DPM exposure of the miners. It will also contribute to the development of a more cost-effective ventilation based on the real time measurement of particulate loading by this instrument to protect mineworkers.

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