

Effects of Diesel Exhaust Aftertreatment Devices on Concentrations and Size Distribution of Aerosols in Underground Mine Air

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Three types of uncatalyzed diesel particulate filter (DPF) systems, three types of high-temperature disposable filter elements (DFEs), and one diesel oxidation catalytic converter (DOC) were evaluated in underground mine conditions for their effects on the concentrations and size distributions of diesel aerosols. Those effects were compared with the effects of a standard muffler. The experimental work was conducted directly in an underground environment using a unique diesel laboratory developed in an underground experimental mine. The DPF systems reduced total mass of aerosols in the mine air approximately 10-fold for light-load and 20-fold or more for high-load test conditions. The DFEs offered similar reductions in aerosol mass concentrations. The efficiency of the new DFEs significantly increased with accumulation of operating time and buildup of diesel particulate matter in the porous structure of the filter elements. A single laundering process did not exhibit substantial effects on performance of the filter element. The effectiveness of DPFs and DFEs in removing aerosols by number was strongly influenced by engine operating mode. The concentrations of nucleation mode aerosols in the mine air were found to be substantially higher for both DPFs and DFEs when the engine was operated at high-load modes than at low-load modes. The effects of the DOC on mass and number concentrations of aerosols in mine air were relatively minor when compared to those of the DPF and DFE systems.

Introduction

In recent years, health issues associated with exposure to diesel particulate matter (DPM) and other, primarily combustion-generated, nano and ultrafine aerosols have received substantial attention from the public, government agencies, and in academia. Long-term exposure to combustion-related fine particulate pollution is perceived as an important risk factor for cardiopulmonary and lung cancer mortality (1).

Extensive utilization of diesel-powered equipment by the mining industry makes the reduction of underground miners'

exposure to particulate matter and gaseous emissions from diesel-powered equipment a major challenge. Techniques for reducing worker exposure typically involve one or more of the following methods; improvements in mine ventilation, the curtailment of DPM and toxic gaseous emissions through improved engine maintenance, exhaust aftertreatment technologies, and the use of alternative fuels.

Diesel particulate filter (DPF) systems are recognized as an effective technology for removing DPM from the exhaust of diesel-powered equipment (2–4). Diesel exhaust filtration systems with disposable filter elements (DFEs) have been extensively used to control DPM emissions from permissible heavy-duty diesel-powered coal mining equipment since the early 1990s (5). Various models of DPFs and DFEs are currently accepted by the U.S. Mine Safety and Health Administration (MSHA) for controlling DPM emissions from underground coal mining equipment (6–9). Although a limited number of underground mining vehicles in operation are currently retrofitted with DPF and DFE systems it can be expected that this number will increase with advancements in engine, DPF and DFE technologies.

The findings of laboratory (10) and field studies (3, 4, 11) indicate that the introduction of various diesel exhaust aftertreatment technologies dramatically changes the physical and chemical properties of diesel aerosols and potentially changes their associated health effects. Growing evidence suggests that particle number, surface area, size, or perhaps some associated structural properties may affect nanoparticle toxicity, when compared with larger respirable particles of the same composition (12).

This paper summarizes the results of a study conducted to evaluate the effects that several types of DPFs, DFEs, and a DOC have on the concentration and size distribution of diesel aerosols in an underground mine. Previous studies on size-resolved characterization of diesel aerosols have typically been performed in well-controlled and ideal laboratory environments (13–16), on roads (11, 16), and in tunnels (17, 18). Since the size and concentration of diesel aerosol and semivolatile materials emitted by diesel engines have been shown to be strongly influenced by a number of complex processes (19), the National Institute for Occupational Safety and Health (NIOSH) chose to assess the aforementioned effects in an underground mine setting. In order to achieve this goal, while still preserving relative precision, a unique diesel laboratory was developed in a nonoperational limestone mine; the NIOSH Lake Lynn Experimental Mine (LLEM), near Fairchance, PA (20). Although the measurements were taken underground, the findings should also be applicable to other occupational settings where workers are performing their duties in proximity to diesel-powered equipment.

Experimental Section

A schematic of the laboratory layout is shown in Figure 1. The D-drift is approximately 530 m (1750 ft) long, 6 m (20 ft) wide, and 2 m (7 ft) high. The major components of the laboratory are an engine/dynamometer system, three sampling and measurement stations, and a ventilation measurement and control system.

The Isuzu C240, one of the most popular light-duty engines in U.S. underground coal mines, was operated at four steady-state engine operating modes (Table 1).

The modes were selected to cover a wide range of engine operating parameters such as exhaust temperatures and emission rates. The average exhaust temperatures at the inlet

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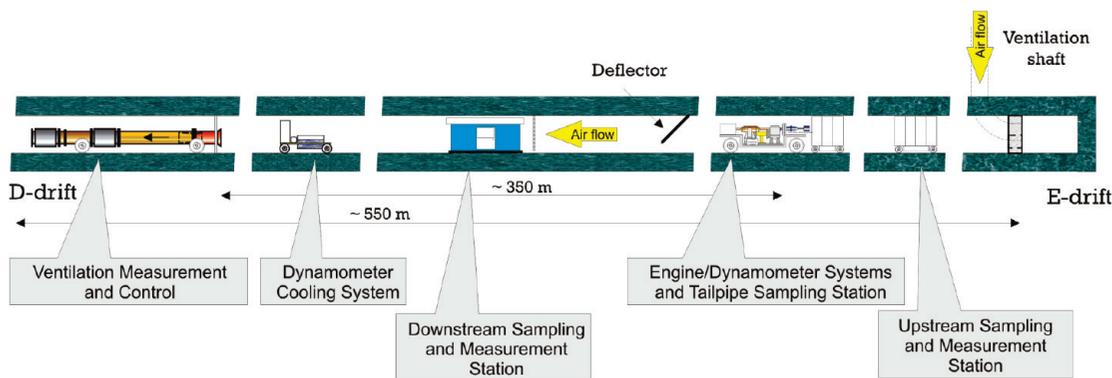


FIGURE 1. NIOSH Diesel Laboratory in D-drift of LLEM (not to scale).

TABLE 1. Test Modes

| mode | description | engine speed (rpm) | torque (Nm) | power (kW) |
|------|------------------------------|--------------------|-------------|------------|
| R50 | rated speed 50% load | 2950 | 55.6 | 17.2 |
| R100 | rated speed 100% load | 2950 | 111.2 | 34.3 |
| I50 | intermediate speed 50% load | 2100 | 69.1 | 14.9 |
| I100 | intermediate speed 100% load | 2100 | 136.9 | 30.6 |

and outlet of the tested DPFs, DFEs, DOC, and muffler observed for test modes are summarized in Table 2.

Fresh air was supplied to the underground facility via a ventilation shaft located in E-drift (Figure 1). A series 1000 model 23017-3450 Axivane fan (Joy Technologies Inc., Franklin, PA) and a subsonic Venturi meter (Primary Flow Signal Inc., Tulsa, OK) were used to maintain and measure constant flow of fresh air through the drift throughout the tests. The measurements showed the average volumetric flow rate of $5.687 \pm 0.047 \text{ m}^3/\text{s}$ ($12050 \pm 100 \text{ ft}^3/\text{min}$). The very low test-to-test variability in flow rate (0.86%) eliminated the need for normalization of the data with respect to it. The average exhaust dilution ratios for R50, R100, I50, and I100 engine operating modes were calculated to be 148, 149, 186, and 188, respectively. The average ambient temperatures upstream of the engine were between 10.5 and 18.7 °C. The corresponding average ambient temperatures at the downstream measurement station ranged between 14.7 and 22.3 °C.

Exhaust Aftertreatment Technologies. Three types of DPF systems (representative of that which is currently available to the underground mining industry) were evaluated in this study:

1. Catalytic Exhaust Products (CEP), Toronto, ON, model 912-SXT with uncatalyzed Corning EX-80 Cordierite element (31 cell per cm^2 and 0.3 mm wall thickness) (Cordierite DPF).
2. DCL International Inc., Concord, ON, model Minex Sootfilter 5.66 × 10 with uncatalyzed Ibsiden silicon carbide element (31 cell per cm^2 and 0.36 mm wall thickness) (SiC DPF).
3. Mann+Hummel GMBH, Speyer, Germany, model SMF-AR with uncatalyzed sintered metal element (10 μm mean pore size, 45% porosity, and 0.38 mm wall thickness) used with Satacen 3 fuel additive (SM DPF).

The DPF systems with Cordierite and silicon carbide wall flow monoliths similar to those from CEP and DCL meet MSHA criteria, and they are listed as 85 and 87% efficient, respectively, in the removal of total DPM mass (6). The SM DPF system was recently added to the list as 99% efficient when used with ultra low sulfur fuel.

All of the DPF systems were installed at an identical location in the exhaust system for their respective tests. The

maximum allowable pressure drop across the DPF systems for all engine modes and DPM loads was set to 14.9 kPa (60" H_2O). Consequently, the DPF systems with a $25.4 \times 30.5 \text{ cm}$ ($10 \times 12 \text{ in}$) Cordierite element and with a 2.75 m^2 sintered metal element were evaluated as single element units, while the SiC DPF system was evaluated with two $14.4 \times 25.4 \text{ cm}$ ($5.66 \times 10 \text{ in}$) elements mounted in parallel. A butterfly valve was installed in the exhaust pipe between the engine and DOC (or muffler) and was used during the DOC and muffler tests to generate pressure drops comparable to those observed for the corresponding DPF and DFE tests. Prior to the start of the first evaluation runs, the DPFs were "de-greened" and fully regenerated using a CombiClean automatic cleaning station from Engine Control Systems, Newmarket, ON.

Three types of high-temperature DFEs with synthetic filtration media were tested using a DFE system consisting of a custom filter housing and an air-to-air heat exchanger designed and built by Mac's Mining Repair Service (Huntington, UT). The heat exchanger was installed between the engine and the DFE housing to cool exhaust below 343 °C (650 F). The following DFEs were evaluated:

1. Donaldson Company, Minneapolis, MN, model P604516 (DFE-A);
2. A Laundered DFE, identical to DFE-A, but laundered by Mac's Mining Repair Service, Huntington, UT, following standard protocols (LDFE-A);
3. FST Systems Corporation, Price, UT, model FST-115-26 (DFE-B).

DFE-A and DFE-B meet MSHA criteria for permissible and nonpermissible applications, and they are listed as 83 and 80% efficient, respectively, in the removal of total DPM (6). The laundering process is used by some coal operators to extend the life cycle of the high-temperature DFEs. The laundered version of DFE-A (LDFE-A) tested in this study was laundered only once.

Two brand new elements (DFE-A and DFE-B) and the laundered element (LDFE-A) were conditioned prior to the 4-mode tests through 13 and 6 h of operation at R50 mode, respectively. The results of measurements performed during this conditioning period were used to assess the effects the filter element loading had on the effectiveness of DFEs.

The observed pressure drops across the DFEs varied with engine operating mode and DPM load, but they were well below the manufacturer-recommended maximum limit of 14.95 kPa (60" H_2O) for all test modes.

A DOC used in this study was manufactured by Engine Control Systems Ltd., Newmarket, ON (model A16-0130) with Cordierite substrate and proprietary catalyst formulation. The baseline measurements were made with a generic muffler installed in place of the DPF systems and DOC.

Fuel. Single-consignment ultralow sulfur diesel fuel (11 ppm S by weight) supplied by Guttman Oil (Belle Vernon, PA) was used for all tests in this study (see Supporting

TABLE 2. Average Exhaust Temperatures at the Inlet and Outlet of the Devices

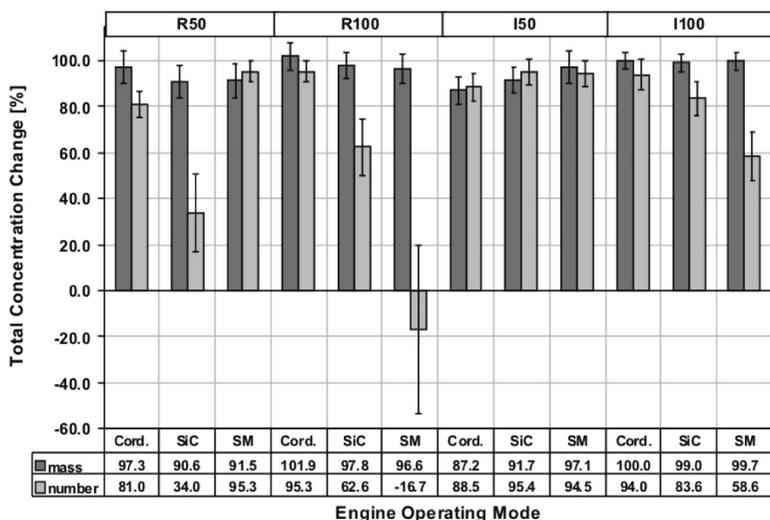
| mode | DPFs | | DFEs | | DOC | | Muffler | |
|------|------------|-------------|------------|-------------|------------|-------------|------------|-------------|
| | inlet (°C) | outlet (°C) |
| R50 | 310 ± 5 | 257 ± 13 | 203 ± 6 | 154 ± 4 | 298 ± 2 | 274 ± 2 | 302 ± 1 | 255 ± 1 |
| R100 | 535 ± 10 | 427 ± 17 | 328 ± 0 | 238 ± 4 | 500 ± 2 | 455 ± 2 | 536 ± 4 | 440 ± 2 |
| I50 | 256 ± 5 | 213 ± 14 | 157 ± 4 | 120 ± 9 | 249 ± 1 | 226 ± 1 | 253 ± 1 | 213 ± 1 |
| I100 | 532 ± 9 | 424 ± 21 | 313 ± 6 | 230 ± 10 | 518 ± 6 | 460 ± 5 | 550 ± 2 | 451 ± 2 |

Information (SI) Table 1 document for the properties). For the majority of the tests, the fuel was supplied to the engine from a 200 L main fuel tank (Rohmac Inc., Mt. Storm, WV). The exceptions were the tests involving the SM DPF system, which was evaluated using fuel from the same batch, but treated with fuel additive from Innospec Limited, Cheshire, United Kingdom, known under the name of Satacen 3. It is important to note that, at the time of testing, this additive was not EPA registered and therefore it was not approved for use in U.S. underground mines. The additive was premixed with the fuel at approximately 0.677 mL per liter of diesel fuel. The additive-treated fuel was supplied to the engine from a designated 45 L fuel tank. Procedures were followed to avoid cross-contamination of the fuels.

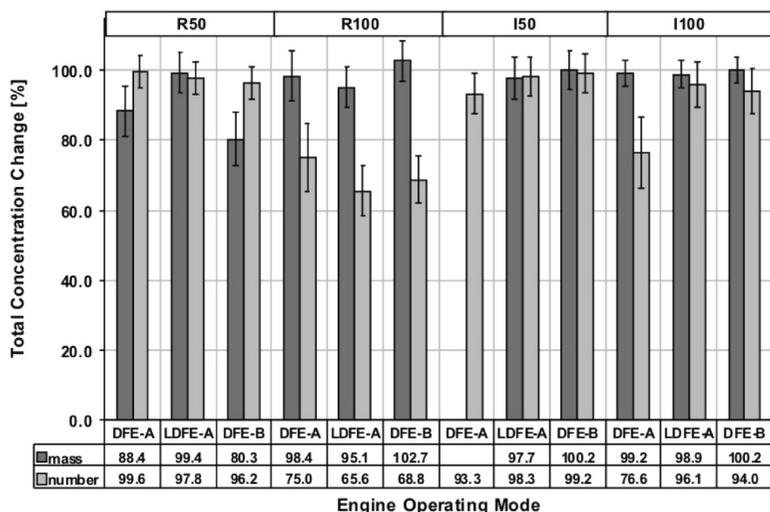
Aerosol Measurements. The effects of the high temperature DFEs and the muffler on aerosols were established by examining results of measurements conducted at two

measurement stations established in the D-drift. A downstream station was located about 60 m (197 ft) downwind of the dynamometer, and an upstream ambient monitoring station was located approximately 60 m (197 ft) upwind of the dynamometer and in relatively clean background air. The downstream sampling location was selected to allow for sufficient residence time for initial formation and transformation of diesel aerosols (21) and full mixing in the drift. Background corrections were made by subtracting the results of measurements performed at the upstream station from the corresponding results obtained at the downstream station.

Measurements were initiated at the end of the initial hour of each test (which was dedicated to achieving thermodynamic and concentration equilibriums). The results in this report are based on the average data obtained over the last hour of each test. The purpose of this approach was to allow



(a)



(b)

FIGURE 2. Percentage of change in total mass (TEOM) and number (SMPS) concentrations for (a) DPFs and (b) DFEs (reduction is positive number).

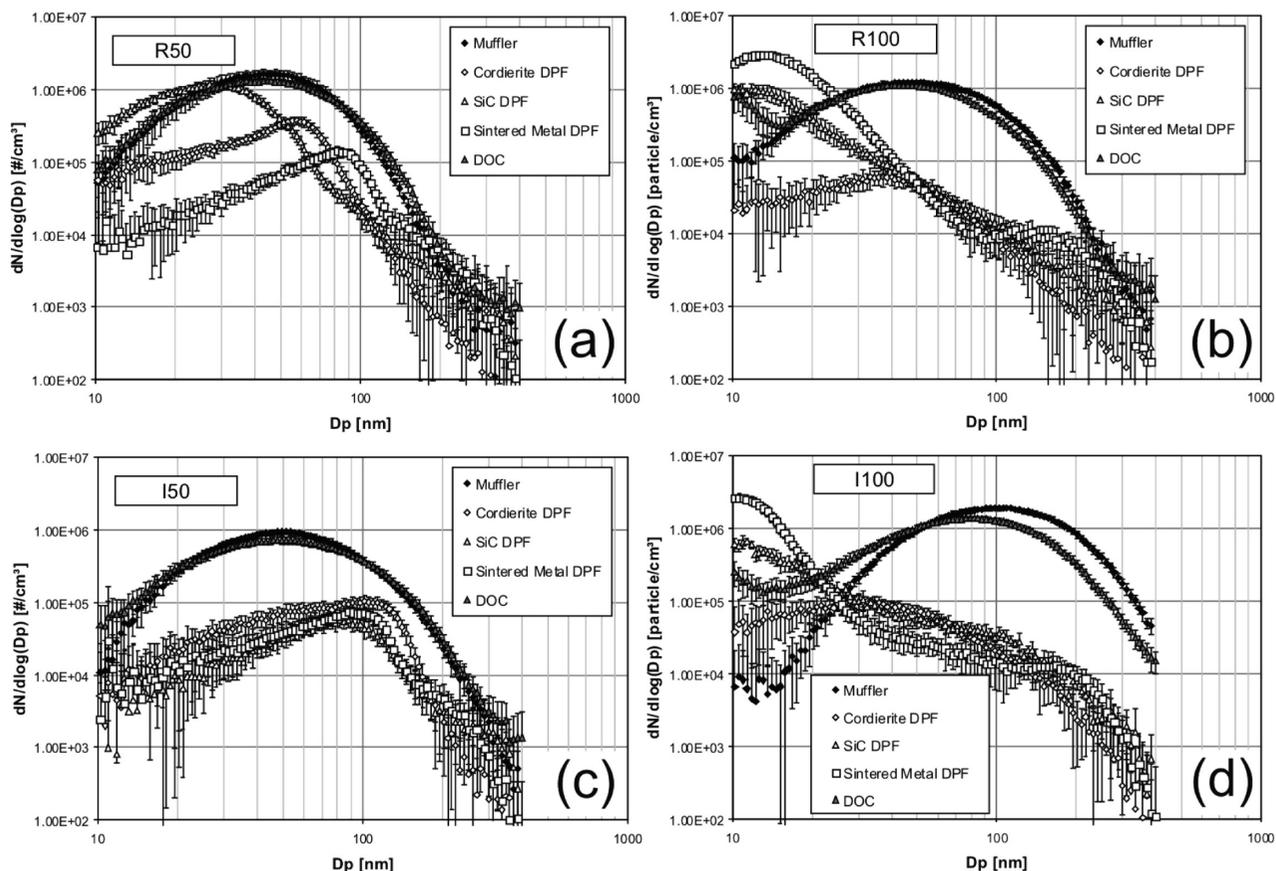


FIGURE 3. Size distributions for the DPFs and the DOC at: (a) R50, (b) R100, (c) I50, and (d) I100 modes.

enough time for DPM to accumulate within the pore structure of the DPFs and DFEs so that the aftertreatment devices were operating at maximum efficiencies during the test.

A tapered element oscillating microbalance (TEOM) series 1400a ambient particulate monitor from Thermo Scientific (Franklin, MA) was used at the downstream station, and a second was used at the upstream station to measure total DPM mass. A 10 mm Dorr-Oliver cyclone followed by a DPM cassette (SKC, Eighty Four, PA) with its collection filter removed were used to preclassify aerosols entering the TEOM, allowing only particles with an average aerodynamic diameter ($_{50}d_{ae}$) smaller than $0.82 \mu\text{m}$ to reach the TEOM. The inlet to the instrument was heated to $50 \text{ }^\circ\text{C}$ following accepted standard practices used to reduce the interference from particle bound water and to minimize thermal expansion of the tapered element.

A scanning mobility particle sizer (SMPS) (22) from TSI (St. Paul, MN) was used at the downstream station, and another was used at the upstream station to measure size distribution and number concentrations of aerosols between 10 and 408 nm. The SMPS at the downstream station was configured with an electrostatic classifier (EC) model 3080 L and a condensation particle counter (CPC) model 3025A. The SMPS at the upstream station consisted of an EC model 3080 L and a CPC model 3776.

Results and Discussion

Effects on Concentrations of Aerosols. The total aerosol mass and number concentrations were measured at the downstream and upstream sampling locations at prevailing ventilation conditions. The results of the measurements performed during the last hour of each test were used to calculate representative average values (see SI Table 2). The average efficiencies of each aftertreatment device in reducing DPM mass and number concentrations were calculated by

comparing the average concentrations measured for the device with those observed for the muffler at the same engine mode (Figure 2). The results are background corrected. The error bars represent one standard deviation of the mean.

All three DPFs reduced aerosol mass concentrations by approximately 10-fold for R50 and I50, and by more than 20-fold for high-emissions R100, and by approximately 100-fold for I100 conditions (Figure 2a). For R100, I50, and I100 modes the DFEs reduced the aerosol mass concentrations by 20-fold or more. At R50 the reductions were less than 10-fold for both DFE-A and DFE-B (Figure 2b).

The engine operating modes were found to have a more pronounced effect on the number than on the mass concentrations of aerosols. The effects of DPFs ranged from 20-fold reductions to slight increases in total number concentrations (Figure 2a). For the light load modes, R50 and I50, the DFEs reduced total number concentration between 93.3 and 99.6%. Significantly lower number reductions, ranging from 65.5 to 75.0%, were observed for all DFEs at R100 and for DFE-A at I100 mode. The effectiveness of LDPE-A in the removal of aerosol mass and number was found to be comparable to that of a new DFE-A, indicating that a single laundering resulted in no substantial reduction in the performance of DFE-A.

A series of tests was conducted in order to assess the effects of DFE loading on performance of the new and laundered DFE elements. Once a DFE was installed, the engine was operated at R50 mode for a period of 13 h (DFE-A and DFE-B) or 6 h (LDPE-A) and aerosol data were collected during those periods. All SMPS measurements were initiated after a one-hour equilibration time. With accumulation of operating time and buildup of DPM within the pore structure of the DFEs the total number concentration of aerosols gradually decreased over the first several hours of operation. The corresponding efficiencies of the DFE-A and

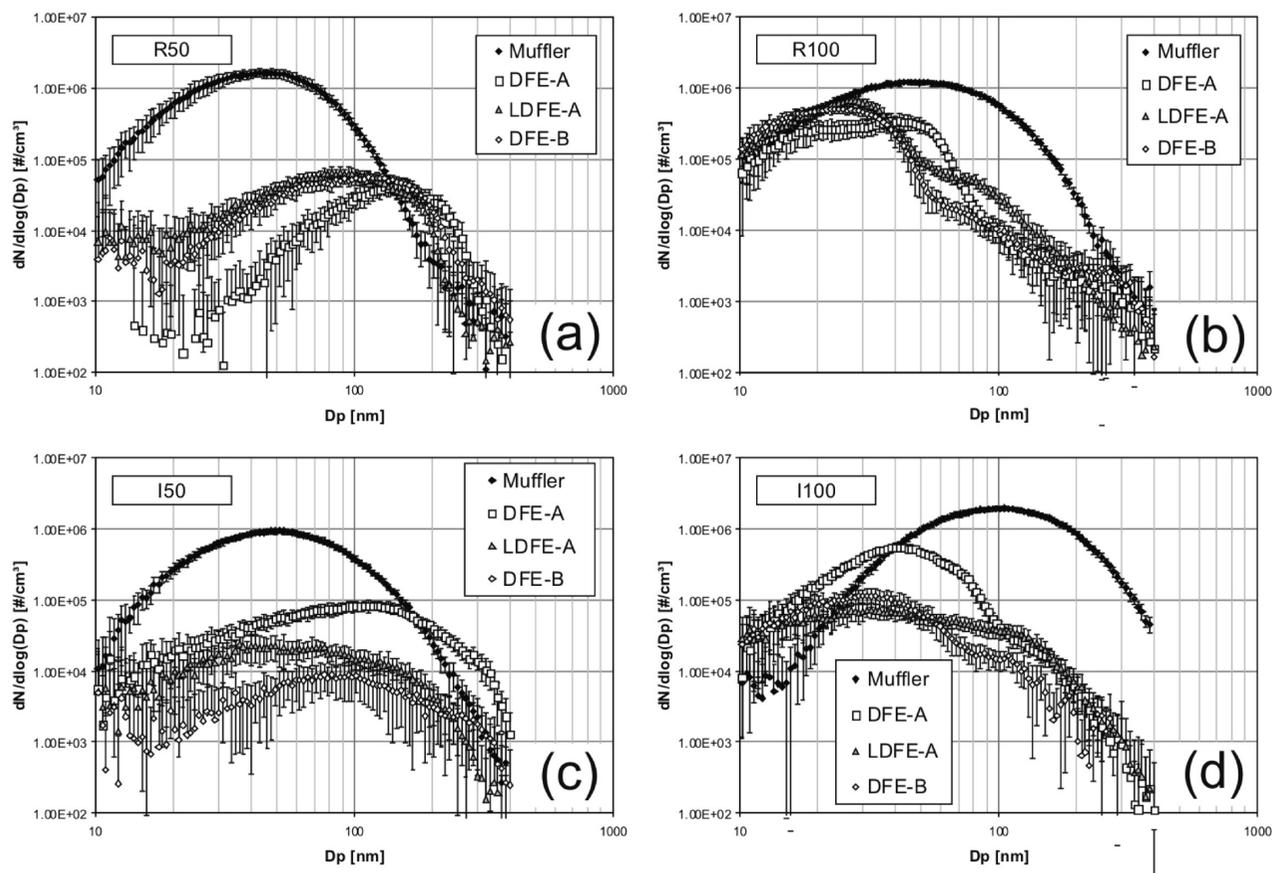


FIGURE 4. Size distributions for DFEs: (a) R50, (b) R100, (c) I50, and (d) I100 mode.

DFE-B increased over the test periods from 83.0 to 99.6 and for 83.0 to 98.0%, respectively. It appears that tested DFEs asymptotically approached but never reached their terminal efficiency over the test periods.

When compared with DPFs and DFEs, the DOC produced relatively modest effects on aerosol mass and number concentrations (see SI Table 2). The effects of the tested DOC on total mass and number concentrations and size distributions of aerosols were found to be influenced strongly by engine operating mode. Substantial data variability and relatively modest reductions resulted in higher uncertainty for DOC results than for the other technologies tested. The most substantial reductions of 42% in mass and 24% in number concentrations were found for the I100 mode.

Effects on Size Distribution of Diesel Aerosols. The results of size distribution measurements performed with an SMPS at the downstream station are summarized in Figures 3 and 4. The presented distributions are not corrected for dilution ratio or for background concentrations.

For all test modes during muffler and DOC tests, the majority of aerosols were found to be in a single agglomeration mode. Although the corresponding aerosol size distributions for the muffler and the DOC were found to be similar, the geometric mean diameters were consistently found to be lower for the DOC cases (40.2 nm vs 40.6 nm for R50, 43.4 nm vs 45.2 nm for R100, 46.7 nm vs 49.6 nm for I50, and 73.8 nm vs 95.5 nm for I100). This probably resulted from a modest catalytic oxidation of the organic fraction of particulate matter, which appeared to be the most effective at I100 conditions. Slightly elevated concentrations of nucleation aerosols were found only when the engine fitted with the DOC was operated at I100 mode. This is consistent with observations made by Maricq and coauthors (10).

For the DPFs, the majority of aerosol size distributions were found to be bimodal with pronounced nucleation ($_{50}d_{em}$

< 50 nm) and accumulation modes ($_{50}d_{em}$ > 50 nm). The aerosol concentrations were found to be primarily in the nucleation mode when the engine was operated at the heavy load and high exhaust temperature conditions, R100 (Figure 3b) and I100 (Figure 3d), and in the accumulation mode when the engine was operated in the light load and lower temperature conditions I50 (Figure 3c). All three types of DPFs were found to be similarly effective in reducing the accumulation mode aerosols from mine air for all test conditions. Although the concentrations of nucleation mode aerosols at R100 and I100 conditions were found to be substantially different between the DPF tests, due to experimental limitations it was not possible to differentiate the effects of the filtration media and a number of the other parameters.

High number concentrations of nucleation mode aerosols in the mine air during the evaluation of SiC and SM DPFs at R100 (Figure 3b), I100 (Figure 3d), and even R50 (Figure 3a) modes, resulted in low or even negative number efficiencies calculated for those modes (Figure 2a). However, for I50 mode, the DPF systems were found to be almost equally effective in removing number and mass of aerosols from mine air (Figure 2a). This can be attributed to low concentrations of nucleation mode aerosols in downstream mine air during the I50 mode tests (Figure 3c).

The size distributions for all three types of DFEs were similar in shape (Figure 4) with aerosols found in both nucleation and accumulation modes. At high load modes, the concentrations of aerosols in the nucleation modes were higher than those in the accumulation modes, which explain the modest total aerosol number concentration reductions observed for all DFEs at those modes. For all test conditions and for all three DPFs, the peak concentrations of nucleation aerosols did not substantially exceed corresponding concentrations measured during the evaluation of the

muffler. The size distributions for LDFA-A were found to be comparable to those of a new DFE-A, showing that a single laundering did not substantially affect performance of DFE-A.

Figure 4a and c show that the size distributions measured for DFE-A and DFE-B tests at R50 and during DFE-A at I50 had two accumulation modes. The relatively high concentration of secondary accumulation mode aerosols with $d_{50} > 100$ nm was attributed to background. The presence of those aerosols can explain the modest total mass reductions found for those conditions (Figure 2a).

The results of size distribution and concentration measurements for DPFs and DFEs, but not for the muffler and DOC, show a positive correlation between concentrations of nucleation mode aerosols in the mine air and exhaust temperatures (Table 2). The concentrations of nucleation mode aerosols were found to be substantially higher when the DPFs were examined at the heavy load and higher exhaust temperature conditions, R100 (Figures 3b and 4b) and I100 (Figures 3d and 4d), than at the lighter load and lower exhaust temperature conditions, R50 (Figure 3a and b) and I50 (Figures 3c and 4c). Substantially lower concentrations of nucleation mode aerosols were observed when the DFEs were examined at high load modes. It is important to note that, due to the effects of the heat exchanger, the DFEs were operated at much lower corresponding exhaust temperatures (Table 2).

The rate of nucleation is likely related to the propensity for hot hydrocarbon-rich gases to pass through the DPFs and DFEs under high exhaust temperature operating conditions and subsequently nucleate downstream as the exhaust cools. Due to the high efficiency in removing the accumulation mode aerosols, the DPFs and DFEs reduce the potential adsorption sites and scavenging of particle precursors, thus promoting heterogeneous and homogeneous nucleation, while at the same time decreasing coagulation of nucleation mode aerosols with accumulation mode aerosols (23, 24).

It is important to note that the reported number efficiencies (Figure 2) were calculated using aerosol counts for the SMPS configuration measurement range, 10–408 nm. In several cases where the size distribution measurements revealed substantial concentrations of nucleation mode aerosols (Figures 3 and 4), one could construe that the concentration of aerosols with diameter smaller than 10 nm contributed significantly to the total number of aerosols. Therefore, if aerosols with diameters smaller than 10 nm were considered, the theoretical increases in number of aerosols in those cases would be substantially higher than those reported in Figure 2.

Variability in the quality of background air (see SI Figures 1 and 2) introduced a higher level of uncertainty into mass and number measurements during the evaluation of very efficient DPF and DFE systems. In those cases, the downstream mass concentrations were found to be comparable to the upstream concentrations and even slight test-to-test variations in concentrations or size distributions of aerosols at the upstream sampling station were found to have a major impact on aerosol mass results. The certainty of number concentration measurements was affected by temporal fluctuations in concentrations of nucleation mode aerosols and background concentrations (Figures 3 and 4). Since the upstream number concentrations were on average between 0.2 and 20.2% of the corresponding downstream concentrations, the effects of background were somewhat less pronounced on number than on mass results. Lastly, changes in the performance of the DPF and DFE systems with accumulation of DPM in filter media over the one-hour averaging period contributed to the overall uncertainty of both mass and number measurements. This raises the more general question of test length and amount of device

conditioning needed to properly evaluate such filtration technologies in both laboratory and field environments.

This study demonstrated the potential of certain diesel emissions control devices to reduce mass and, in the majority of cases, number concentrations of diesel aerosols in occupational settings such as underground mines. The results showed evidence of the formation of high concentrations of nucleation mode aerosols when DPFs and DFEs were used in the exhaust system of engines operated at high exhaust temperature conditions. Future work is necessary to fully understand how these changes in the characteristics of diesel emissions may impact the toxicological properties of these aerosols and the potential health risks associated with worker exposure to DPM.

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Supporting Information Available

Additional information available. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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