

Laboratory demonstration of DPM mass removal from an exhaust stream by fog drops

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Abstract ■ Diesel particulate matter (DPM) is a major occupational health hazard in underground mine environments, even with available abatement technologies. Prior work has demonstrated that micrometer-scale water droplets, or “fog,” may be effective in removing significant number fractions of DPM from a conditioned exhaust stream. Here, a series of laboratory experiments are described which demonstrate that such a treatment can remove significant DPM mass from an unconditioned stream. The removal mechanism involved coagulation of the DPM and drops. The fog treatment removed about 45 percent more DPM mass on average than no treatment. Varying engine load, flow rate and fog droplet number density did not significantly affect the results under the range of conditions tested.

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Introduction

Diesel engines have seen widespread use for well over a century due to their relatively high thermal efficiency and fuel economy (Heywood, 1988). Recently, however, the adverse health risks of diesel exhaust have become increasingly clear. The term diesel particulate matter (DPM) is used to refer to the solid components of diesel exhaust, which are an ultrafine mixture of elemental and organic carbon (EC and OC) and minor constituents including sulfates and metal

ash (Kittelson, 1997). DPM is generally considered to occur almost entirely in the submicrometer range. It is classified as a carcinogen (Occupational Safety and Health Administration, 2013), and epidemiological studies have demonstrated a positive correlation between long-term exposure to DPM and other combustion-related fine particulates and increased cardiovascular and pulmonary diseases (Pope et al., 2002; McDonald et al., 2011). Many of the risks of diesel exhaust are associated with the physical and chemical properties of exhaust components (Heywood, 1988; El-Shobokshy, 1994; Kittelson, 1997). Exposures are generally measured and regulated on the basis of mass concentration. However, DPM number density and particle size are increasingly recognized as critical factors in terms of health outcomes (Bugarski et al., 2012; Kittelson, 1997; Occupational Safety and Health Administration, 2013; Pope et al., 2002).

Diesel engines operate in relatively fuel-rich/oxygen-lean conditions and are characterized by high emissions of particulates relative to those from spark-ignition engines (El-Shobokshy, 1994; Kittelson, 1997; Fiebig et al., 2014). Emissions from large equipment such as that used in mining ap-

plications typically range from 10^7 to 10^9 DPM particles per cubic centimeter (Kittelson, 1997). The physical and chemical properties of DPM vary with the type of engine, fuel and operating conditions such as loading, which is a function of torque and rotational speed (El-Shobokshy, 1994; Kittelson, 1997; McDonald et al., 2011; Bugarski et al., 2010; Huang et al., 2015). Loading is a particularly important factor with respect to DPM toxicity (McDonald et al., 2011; Stevens et al., 2009; McDonald et al., 2004) and the effectiveness of after-treatment technologies (El-Shobokshy, 1994; Kittelson, 1997; An et al., 2012). Engine load alone can affect the EC/OC ratio, and the size distribution and number density of DPM. Light loads generally favor the formation of OC and small particles. As load is increased, the volatiles are oxidized, leading to larger soot particles (EC) but lower total number density of DPM. With further loading, the formation of soot offsets the decrease in volatiles, resulting in increased DPM mean size and number density (Kittelson, 1997).

A significant body of work has been devoted to the development of DPM-reducing technologies, including after-treatments like oxidation catalysis and

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particulate filters (Bugarski et al., 2009). However, operational constraints and/or economics can hinder implementation of such technologies (Bugarski et al., 2009). Even with them, occupational exposures are still a serious concern in many underground mines, where large fleets of diesel equipment are operating in confined spaces. Moreover, as is the case for exposure monitoring, evaluative criteria for DPM abatement technologies have generally been on a mass basis. From a health perspective, new technologies should ideally address both mass and number reductions.

A limited amount of available research has suggested that DPM abatement through water treatment may hold promise. For example, some disposable filter element (DFE) systems designed for coal mining equipment have included on-board water bath conditioners for cooling exhaust upstream of the filter (Bugarski et al., 2011), where the water bath itself can yield a DPM mass removal of up to 35 percent (Bugarski et al., 2012). Moreover, several studies have considered DPM scavenging by water droplets (D’Addio et al., 2013; Ha et al., 2009), and there is ample literature aimed at the use of water sprays to capture airborne particulates more broadly (Cheng, 1973, Kim et al., 2001, Koo, Hong and Shin, 2010; Ran, Saylor and Holt, 2014). For the case of DPM, both the ultrafine particle size and exhaust conditions, such as flow rate and temperature, present challenges to treatment design.

Our prior work has suggested that micrometer-scale water droplets, or “fog,” can be effectively used to scavenge DPM from an airstream (Rojas-Mendoza et al., 2015, 2017). In one set of experiments, diluted and electrically neutralized engine exhaust was subjected to a fog treatment to promote coagulation (Rojas-Mendoza et al., 2017). Measurements upstream and downstream of the treatment showed that the fog removed about 45 percent more DPM on a number basis from the exhaust stream than no treatment. Importantly, the fog had similar effects over the entire DPM size range observed: 25 to 100 nm. Only a limited number of mass-based results were collected in that study. A two-step mechanism of water drop-DPM coagulation followed by settling was proposed to explain the results. In that case, the size and number density of DPM and drops are important variables, as they dictate the rate at which coagulation oc-

curs. The system residence time is also key, as it dictates the window of opportunity for both coagulation and settling. For fixed-volume systems, flow rate controls the residence time and also turbulence, which may in turn influence additional DPM removal mechanisms, such as impaction of particles with system surfaces.

Expanding upon the abovementioned prior work, the objective here was to demonstrate DPM mass removal from an unconditioned exhaust stream using fog droplets. Experiments were performed to specifically investigate the effects of (1) engine loading, (2) system flow rate and residence time, and (3) number of fogging devices used.

Experimental procedure

A schematic of the experimental apparatus is presented in Fig. 1. Diesel exhaust was generated and brought into contact with fog droplets under different experimental conditions. The percent of mass removed by the treatment was quantified by:

$$L_M(\%) = \left(\frac{M_U - M_D}{M_U} \right) \times 100 \quad (1)$$

where M_U and M_D correspond to the mass of DPM collected simultaneously upstream, or location A, and downstream, or location B, of the treatment.

A Kubota EA330-E4-NB1 Engine (Kubota Engine America, Lincolnshire, IL) with maximum output of 5,150 W and maximum speed of 3,000 rpm was used as the DPM source. The engine was coupled to a portable belt-driven generator with maximum output of 2,900 W. Load was applied to the engine using incandescent light bulbs, each rated at 72 W.

A diaphragm pump was used to take a bleed off of approximately 1 percent of the total diesel exhaust, which cooled almost immediately to room temperature. However, the exhaust was not diluted and was not electrically neutralized as in Rojas-Mendoza et al. (2017). Exhaust flow was measured using a high precision rotameter, and was directed to the treatment area of the experimental setup (within the dashed line in Fig. 1). The primary component of the treatment area was a fogging chamber, with sampling lines located just upstream and downstream. The fogging chamber was a Plexiglas cube with each side measuring 20.3 cm (8 in.). A water pool was located within the fogging chamber that

Figure 1
Experimental apparatus.

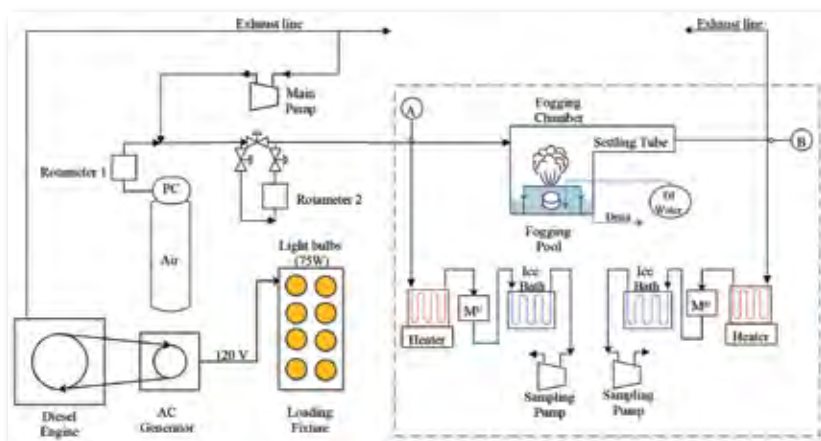


Table 1

Experimental conditions for each engine run.

Engine run	Flow condition	Load condition	Sequence of fog treatment conditions (device no.)				Sampling time (min)	Engine rpm
			1 st	2 nd	3 rd	4 th		
1	HF	HL	ON (1)	OFF	ON (1)	OFF*	60	2,216
2	LF	HL	ON (1)	OFF	ON (1)	OFF	60	2,195
3	HF	LL	ON (1)	OFF	ON (1)	OFF	60	2,205
4	LF	LL	ON (1)	OFF	ON (1)	OFF	60	2,190
5	LF	HL	ON (1)	OFF	ON (2)	OFF	20	2,216
			ON (4)	OFF	ON (4)	OFF		
			ON (1)	OFF	ON (2)	OFF		

*Sample could only be collected for 30 min instead of 60 min.

had a replacement rate of 0.9 L/min (0.2 gpm). This minimized contamination of the water surface from which fog droplets were generated. Deionized water was used in all cases. The remainder of the fogging chamber served as the primary treatment system volume of 5,637 cc (344 cu in.), and included a drain to continually remove water overflow from the water pool.

A cylindrical Alpine FG100 fogging device (Alpine Corp., Commerce, CA), 3.8 cm (1.5 in.) in diameter and 2.5 cm (1 in.) tall, was used to produce water droplets. The exhaust/fog mixture exited the chamber through a 61-cm (24-in.)-long settling tube with inner diameter of 4.45 cm (1.75 in.) and volume of 913 cc (56 cu in.). The fogger used an ultrasonic transducer whose acoustic energy was directed upward through the water to the air-water interface, resulting in the formation of water droplets at that interface. It required 24 W of power and generated water drops at a rate of less than a milliliter per minute. The drops had an estimated mean size of 3.9 μm , with 95 percent between 1.9 and 7.3 μm , and a number density of about 5.0×10^5 drops/cc under the conditions tested. The size of the water droplets was determined by allowing the drops to impact a glass slide, and then by imaging them with a Zeiss Axiovert 200M MAT stereoscope microscope (Carl Zeiss AG, Oberkochen, Germany) coupled with an AxioCam MRc5 digital camera. Images were processed using ImageJ (National Institutes of Health, Bethesda, MA). The number density of droplets was estimated by using the fogging device with a fluorescein solution. The generated fog was passed through a diffusion dryer, and then a TSI 3910 NanoScan SMPS 10-420 nm nanoparticle sizer (TSI Inc., Shoreview, MN) was used to measure the number concentration of particles.

Paired mass samples were collected at locations A and B on polycarbonate filters, 37 mm (1.5 in.) in diameter with pore size of 0.2 μm , in standard two-piece air sampling cassettes. Escort ELF pumps (Zefon International, Ocala, FL) calibrated to a flow rate of 1.7 L/min (0.45 gpm) were used. In order for significant DPM mass to be collected on the hygroscopic polycarbonate filter without stalling or flooding the sampling pump, particularly at location B, the tubing upstream of the sample cassette was immersed in a hot water bath to minimize liquid water accumulation on the filter. The tubing downstream of the cassette was immersed in an

ice bath to promote condensation, which was trapped before air entered the sampling pump. This was done at both locations A and B to be consistent. Filters were weighed before and after sample collection using a Sartorius Cubis MSE6.6S microbalance (Sartorius AG, Göttingen, Germany). Sample moisture was removed by drying the samples at 40 °C (104 °F) for 10 hours. Sample mass was determined simply as the difference between the DPM-loaded dry filter and the clean dry filter prior to sample collection.

Two groups of experiments were performed during five separate engine runs (Table 1). In the first group, consisting of runs 1 to 4, engine load and exhaust flow rate were varied independently using a single fogging device. In the second group, consisting of run 5, fog droplet number density was varied by using different numbers of foggers while keeping the engine load and exhaust flow rate constant. To attain a stable condition, the engine was warmed up for 60 min prior to each run, ultrapure air was purged through the treatment system, and the exhaust bleed off was then directed through the treatment system, and data collection commenced. The fog was alternated between being ON or OFF in each run (Table 1), and DPM mass samples were collected at locations A and B for each instance, such that two sets of ON samples and two sets of OFF samples were obtained under each unique test condition. The 95-percent confidence intervals for the mean L_M values during fog ON or OFF in each group of experiments were calculated using one-way analysis of variance. The confidence intervals for the improvement in removal attributed to the fog, $L_{M-ON} - L_{M-OFF}$, were calculated using the multiple comparison procedure known as the least significant difference method. JMP Pro 13 statistical software (SAS, Cary, NC) was used to do the statistical analysis.

To vary engine load, two modes were used. Low load (LL) was achieved by illuminating zero light bulbs, and high load (HL) was achieved by illuminating 16 bulbs for a total of approximately 1,200 W. Prior to starting experiments and collecting DPM mass samples, the DPM size distribution under each loading condition was characterized. These number-resolved size distributions were obtained at location A using the NanoScan and a TSI 3330 300-10,000 nm optical particle sizer (TSI Inc., Shoreview, MN). Dilution air was necessary in order to obtain accurate number-resolved

measurements so that the number density of particles in the exhaust stream was within the instruments' range. For the low-load condition, a ratio of 3.7 L/min of diesel exhaust to 6.3 L/min of dilution air was used. For the high-load condition, a ratio of 3.1 L/min of diesel exhaust to 6.9 L/min of dilution air was used.

As expected, increased loading resulted in higher DPM number densities and increased geometric mean aerosol diameter. Measurements obtained with the NanoScan are shown in Fig. 2. The average number density and geometric mean diameter were $2.89 \times 10^6 \pm 0.48 \times 10^6$ #/cc and 44.22 ± 1.25 nm, respectively, for the low-load condition and $4.21 \times 10^6 \pm 1.05 \times 10^6$ #/cc and 61.16 ± 5.51 nm, respectively, for the high-load condition. Geometric mean diameter (GMD) was calculated using:

$$GMD = (D_1^{n_1} D_2^{n_2} \dots D_N^{n_N})^{1/N} \quad (2)$$

where D_i is the midpoint particle diameter, n_i is the number concentration of particles in bin i having a midpoint diameter D_i , and N is the total number concentration of particles summed over all intervals. Measurements obtained with the optical particle sizer showed that all of the DPM was smaller than 1,000 nm for both loading conditions, and the average number densities from the optical particle sizer were eight orders of magnitude less for low load and three orders of magnitude less for high load than the number densities measured by the Nanoscan under the same loading conditions. Assuming spherical particles and constant specific gravity across all sizes, then DPM in the optical particle sizer range should only account for about 0.2 and 2.2 percent of the total mass concentration at low and high loads, respectively. For this reason, it was considered insignificant for analysis of the experimental results reported here.

Two different residence times were tested by varying the exhaust flow rate through the fixed treatment system volume. Low flow (LF) mode was achieved with a total flow rate of 8.3 L/min (2.2 gpm), measured at the inlet of the fogging chamber, while high flow (HF) used 18.3 L/min (4.8 gpm). Both flow rates were tested at both engine loads.

The number of fogging devices was also independently varied such that either one, two or four devices were operating simultaneously, in an attempt to vary the water droplet number density (these are the numbers in parentheses in Table 1). Each fogging condition was tested under low-flow, high-load conditions. The mean droplet diameter and droplet number density stated above was determined using a single device, and all of the devices used here came from

the same production batch and were assumed to have similar operating characteristics.

Results and discussion

Average L_M during fog ON and fog OFF conditions were calculated using Eq. (1) for each experiment. The difference between these values is used to define the improvement in DPM removal attributed to the fog treatment. Figure 3 shows the results from all experiments, where each condition is labeled according to the convention "flow condition-load condition-number of fogging devices." For example, LF-LL-1 denotes low flow, low load and one fogger. A maximum improvement in DPM removal of 54.1 ± 9.5 percent was observed under the low-flow, high-load conditions with a single fogging device (LF-HL-1). However, 95-percent confidence intervals overlap between all four of the flow-load conditions investigated. This observation is discussed further below. From Fig. 3, it is also evident that deposition losses, which refer to DPM removal in the experimental system when the fog is off, can vary from engine run to engine run.

Figure 3 additionally shows the effect of varying the number of fogging devices, using the data collected in engine run 5. The experiment using one fogger can be considered a replicate of the low-flow, high-load (LF-HL) condition shown in Fig. 3, and results for overall improvement in DPM removal are in reasonably good agreement: 46.2 ± 10 percent versus 54.1 ± 9.5 percent. The improvement in DPM removal for the case of one, two and four fogging devices are similar, giving essentially the same value within the confidence intervals. The maximum improvement in DPM removal was observed to be 53.2 ± 11.5 percent in the case of four foggers, but again the error bars between all conditions overlapped.

Based on all of the results presented here, it is clear that the fogging treatment did yield a significant improvement versus no treatment in DPM mass removal from the engine exhaust stream in this experimental system. However, differences in removal with varying engine load, flow rate and fog density conditions were subtle, and this observation deserves further analysis. For this, it is assumed that the primary DPM removal mechanism from the experimental system involves a two-step process whereby airborne DPM and fog droplets attach due to thermal coagulation, or Brownian motion, and then the DPM-laden drops are removed from the air by gravitational settling and/or impaction with the surfaces in the system due to inertial effects.

First, the DPM-droplet coagulation rate is considered. Effectively, this is the rate at which DPM particles and droplets

Figure 2

DPM size distribution at high load (HL) and low load (LL). Error bars represent 95-percent confidence intervals.

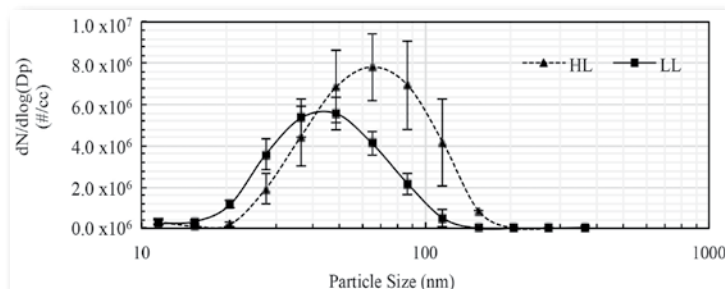
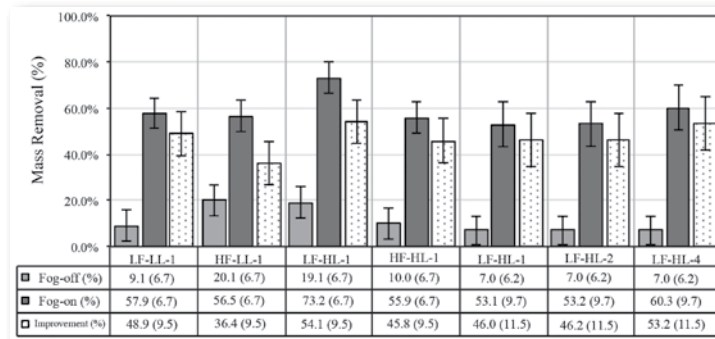


Figure 3

Average L_M values and 95-percent confidence intervals for each treatment condition in experiments varying flow rate (LF or HF), engine load (LL or HL) and fog drop number density (1, 2 or 4 fogging devices). The improvement in L_M attributed to the fog treatment is also shown for each condition.



attach to one another. Attachment can be modeled as the disappearance of airborne DPM from the system as particles are taken up by drops (Hinds, 1999). The water drops are very big and diffuse very slowly, while the much smaller DPM particles diffuse rapidly to the water droplet surfaces. The relative size difference between the drops and DPM drives DPM-drop coagulation to be much more rapid than DPM-DPM or drop-drop coagulation. As developed in detail by Rojas-Mendoza et al. (2017), the coagulation rate can be predicted using the system residence time, and the fog droplet and DPM particle sizes and number densities. Here, the residence time of the fogging chamber and settling tube was calculated to be approximately 47 and 22 s for the low-flow and high-flow conditions, respectively. For the case of a single fogging device, the mean droplet diameter and number density given above were used, and the drop diameters were assumed to be monodisperse. Conservatively, it was also assumed that the droplet diameter remained constant and the number density scaled linearly with the number of foggers used, for example, two and four foggers should provide 10×10^5 and 20×10^5 droplets/cc, respectively. Based on Fig. 2, DPM particle size and number density can also be estimated. For this analysis, the geometric mean size was considered for the five primary size bins, which accounted for 90 and 84 percent of the total DPM particle count in the low-load and high-load conditions, respectively. The bins were 23.7 to 31.6, 31.6 to 42.2, 42.2 to 56.2, 56.2 to 75 and 75 to 100 nm. By considering a limited number of DPM sizes, computation of coagulation rate can be greatly simplified, as each

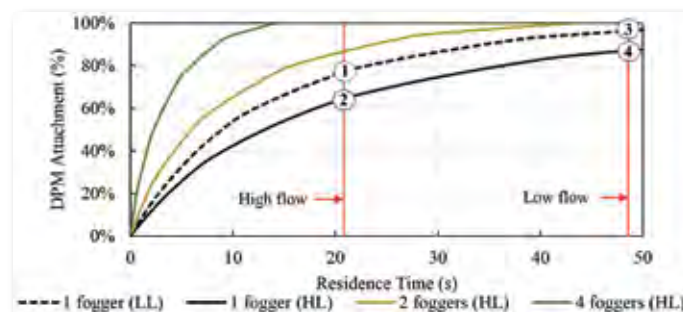
combination of particle size-droplet size must be evaluated.

Using the above estimates and assumptions, and following the analysis detailed in Rojas-Mendoza et al. (2017), the fraction of DPM-drop attachment expected for the experimental conditions tested here is shown in Fig. 4. To examine the influence of engine loading, attachment values can be compared between points 1 and 2 and points 3 and 4. At high flow rate, attachment is expected to be about 77 percent at low load (point 1) and about 65 percent at high load (point 2). By shifting to low flow rate, the expected attachment increases to about 96 percent at low load (point 3) and about 88 percent at high load (point 4). Thus, the variation in engine load tested in the current work is only expected to change coagulation of DPM and fog droplets by about 8 to 12 percent, and the variation in flow rate is expected to change coagulation by about 19 to 23 percent.

Similarly, Fig. 4 illustrates that doubling or quadrupling the fog droplet number density — in low-flow, high-load conditions — should only result in a 12 percent increase in coagulation, and the attachment with a single fogger under these conditions is already predicted to be relatively high, with point 4 at 88 percent attachment. Effectively, the coagulation rate is expected to be fast enough to allow significant attachment within the system residence time tested. At a shorter residence time, the effect of increased fog drop density may have been more apparent. Nevertheless, when considering just the coagulation step of the proposed DPM removal mechanism, it is perhaps not surprising that only modest improvements, if any, were observed upon varying engine load, flow rate or

Figure 4

DPM attachment as a function of residence time for different DPM and water drop number densities.



number of fogging devices within the ranges tested here.

The second step of the proposed mechanism, removal of DPM-laden drops from the air, should also be considered. Flow rate is expected to be the most significant variable in this analysis, as it can influence drop removal in two primary ways: (1) by affecting the residence time, which should have an impact on the fraction of drops being removed due to gravitational settling, and (2) by affecting the turbulence conditions, which should influence the likelihood of impaction of drops with system surfaces.

For 3.9 μm droplets, the settling velocity can be estimated as 1.44×10^{-3} m/s (4.7×10^{-6} ft/sec) using the terminal velocity of a spherical water drop due to gravity, according to:

$$V_T = \frac{g\rho_d d_d^2}{18\eta} \quad (3)$$

where g is the gravitational constant, ρ_d is the density of the water drop, d_d is the drop mean diameter and η is the air viscosity (Hinds, 1999). For the residence times tested here, gravitational settling should thus reduce the number of fog droplets by about 17 and 35 percent at the exit of the settling tube for the high-flow and low-flow conditions, respectively. These estimates are based on a comparison of particle settling time versus system residence time, including the fogging chamber and settling tube. While this analysis indicates that approximately doubling the flow rate should cut the drop removal rate in half, the results shown here are relatively similar for the high-flow and low-flow conditions tested. This suggests that another removal mechanism or other influential factors could be at play, such as differences in particle deposition on system surfaces or water drop stability under different flow conditions.

Conclusions

The primary objective of this work was to demonstrate DPM mass removal from an unconditioned exhaust stream using fog water droplets. The results indicate that the fog treatment had a significant effect, which is attributed to coagulation of the DPM and drops under the experimental conditions tested. Taken together with prior results under similar conditions, which showed reduction in DPM number density (Rojas-Mendoza et al., 2017), it can be concluded that treatment technologies using a DPM-fog coagulation mechanism could be effective on both a mass and number basis. Transferring these results for practical application in a mine setting will, however, require many additional considerations. In the case of a direct-exhaust treatment, flow rate and temperature will pose significant challenges — and may fundamentally alter the DPM removal mechanism. Such an application might seek to take advantage of existing exhaust cooling technologies, like those that have been developed for DFE or other systems, or to employ the fog for both evaporative cooling/condensational growth and coagulation with DPM. A simpler application in terms of design might be for a localized area treatment, such as a scrubber, in which case the ability to generate sufficient concentrations of fog drops may represent a primary challenge. ■

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