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Institution Address:

Regents of the University of Minnesota
200 Oak Street S.E., Suite 450
Minneapolis, MN 55455

Principal Investigator: Peter C. Raynor, Ph.D., Associate Professor

Principal Investigator Contact Information:

University of Minnesota
Division of Environmental Health Sciences
Mayo MC 807, 420 Delaware Street S.E.
Minneapolis, MN 55455
Phone: (612) 625-7135
Email: praynor@umn.edu

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LIST OF TERMS AND ABBREVIATIONS

ANOVA	analysis of variance
APS	Aerodynamic Particle Sizer
B_{SprayOff}	background particle concentration when the spray was not applied
B_{SprayOn}	background particle concentration when the spray was applied
C	Cunningham slip correction factor
C_{SprayOff}	net particle count when the spray was not applied
C_{SprayOn}	net particle count when the spray was applied
CWP	coal workers' pneumoconiosis
D	spray droplet concentration
D_p	particle diameter
e	elementary charge
kV	kilovolts
M	molar
MSHA	Mine Safety and Health Administration
n	number of elementary charges on a particle
NIOSH	National Institute for Occupational Safety and Health
p	p-value
P_{SprayOff}	test particle concentration when the spray was not applied
P_{SprayOn}	test particle concentration when the spray was applied
PEL	permissible exposure limit
PSL	polystyrene latex
REL	recommended exposure limit
SD	standard deviation
Z_p	electrical mobility
η	spray collection efficiency
μ	gas viscosity
μm	micrometer

ABSTRACT

Inhalation exposures to respirable coal dust are a serious health concern for underground coal miners. One method used to limit these exposures is to spray water that contains surfactants to capture the airborne particles near the mining face. The goal of this research was to determine if surfactants that carry an electrical charge can enhance respirable particle capture through electrostatic attraction. In a laboratory setting, tests were conducted to measure the effects of particle diameter, surfactant type and concentration, and particle charge on the capture of respirable particles by surfactant-containing water spray droplets. Polystyrene latex particles with diameters of 0.6, 1.0, or 2.1 μm or coal dust particles with a wide range of diameters were generated in a wind tunnel. Particles were given either an unneutralized, neutralized, net positive, or net negative charge, and then were allowed to be captured as they passed through sprays containing anionic, cationic, or nonionic surfactant, or they moved through the same spray section with the spray turned off. After passing through the spray region, the particles were sampled, charge-separated, and counted at various voltage levels. Comparisons of concentrations with the sprays on and off permitted the calculation of collection efficiency. Overall efficiencies were measured integrated across all charge levels, as well as efficiencies for particles at specific charge levels. The overall collection efficiency rose significantly with increasing particle diameter. Collection efficiencies of 22%, 59%, and 87%, on average, were observed for polystyrene latex particles 0.6, 1.0, and 2.1 μm in diameter, respectively. The combination of surfactant classification and concentration significantly affected both overall spray collection efficiency and collection efficiency for particles with specific charge levels. Anionic (negatively-charged) and cationic (positively-charged) surfactant sprays had the best performance with charged particles having the opposite sign of charge and the worst performance for charged particles holding the same sign of charge. Nonionic surfactant-containing spray removed particles carrying relatively few charges more efficiently than the anionic or cationic surfactant sprays. Particle charge level impacted the spray collection efficiency as highly-charged particles were removed more efficiently than weakly-charged particles. Comparisons of measurements made with the polystyrene latex particles and the coal dust particles showed similar efficiency, indicating that the polystyrene particles are a suitable surrogate for the coal dust particles.

SECTION 1

Significant (Key) Findings

In this study, particle size was the most important determinant of spray collection efficiency for respirable particles. Our data indicated that polystyrene latex (PSL) particles with a diameter larger than 2 μm can be removed effectively by sprays, whether they contain surfactants or not. Therefore, a choice of surfactant to improve dust collection by electrostatic attraction will primarily be important for smaller particles. In addition, the data show clearly that any effects of electrostatic interactions between spray droplets and charged particles are of secondary importance to particle size.

The type of surfactant used in an aqueous spray system significantly impacted collection of respirable particles. Adding ionic surfactant into spray water improved dust capture for highly-charged particles carrying the opposite sign of charge. In other words, sprays containing a cationic surfactant, which carried positive charges, were more effective than other surfactants at collecting particles that held high levels of negative charge. Similarly, sprays containing an anionic surfactant, which carried negative charges, were more effective than other surfactants at collecting particles holding high levels of positive charge. High concentration nonionic surfactant-containing sprays were more effective than the other sprays at capturing particles that held weak charges.

Particle charge level impacted spray collection efficiency significantly. Highly charged particles were removed more efficiently than weakly charged particles by all surfactant sprays. Therefore, in coal mines, highly-charged dust is likely to be collected better by sprays than weakly-charged dust, especially by spray droplets carrying the opposite sign of charge. Knowledge of the charge distribution of an aerosol, or at least the average sign and magnitude of charge on an aerosol, could be an important factor for informing surfactant choice for mine operators.

Measurements showed that PSL particles were a reasonable surrogate for coal dust particles to test the effects of particle charge and surfactant spray characteristics on collection efficiency on a size-by-size basis. This is an important finding because PSL particles are more uniform, more easily generated, and safer and cleaner to work with in a laboratory setting than coal dust. In future laboratory-based research, investigators can simplify their efforts by working with monodisperse PSL particles and have confidence that their findings should apply to polydisperse real-world particles.

Translation of Findings

The central hypothesis of the proposed research is that, for a given respirable coal dust aerosol, a type and concentration of spray surfactant can be selected that will lead to a higher spray collection efficiency than with plain water. Our findings indicate that there is a benefit from using electrical attraction between coal dust particles and surfactant-containing sprays to improve the effectiveness of spray collection. Ultimately, these findings can be used to help underground coal mine operators make informed decisions about selecting the best spray surfactant to maximize capture of the dust generated in their mines. To select an appropriate surfactant, the charge level and sign of respirable particles generated in a mine must be known. The results also indicate that making a surfactant choice merely based on classification (anionic, cationic, nonionic) may not be sufficient. The spray surfactant charge distribution may vary by the particular surfactant and by concentration. Thus, to be able to predict the electrical effect on dust control efficiency and make optimal surfactant choices, we need data for the electrical charge level of dust particles and the electrical charge distribution of the surfactant-containing spray droplets. Then, we can develop expressions to predict spray collection efficiency based on particle and droplet charge levels. To accomplish these goals, further research is required.

To achieve a long-term goal of reducing respirable coal dust concentrations in underground coal mines, the objective of future research should be to develop a selection algorithm for choosing a surfactant to maximize spray collection efficiency in a particular mine based on the charge characteristics of particles in that mine. The hypothesis underlying this approach would be that a specific concentration of a specific surfactant will lead to optimal spray collection efficiency in a particular mine because of the interactions between electrical charges on the surfactant spray droplets and the airborne particles in the mine.

To test this hypothesis, several steps would be required. First, researchers could measure the electrical charge distribution of sprays containing surfactants. Our findings indicate that both surfactant type and

concentration play an essential role in the electrical charge distribution of the spray droplets, which affects particle collection efficiency. The charge distribution as a function of drop size could be measured for a wide variety of surfactant sprays. Second, researchers could measure the electrical charge distribution of respirable particles when surfactant sprays are used and when no spray is used. Our test results suggest that the electrical charge level on particles has significant impact on the collection efficiency. Using PSL particles, tests could be conducted on many combinations of surfactant type and concentration to understand how both size and charge distributions are affected by the presence of a surfactant spray. Third, researchers could use the results of the spray and particle size and charge measurements to develop an algorithm for choosing a surfactant to use in spray water based on the size and charge characteristics of respirable particles from a particular mine. The selection algorithm could work in such a way that, given a mean particle diameter, charge sign, and charge magnitude, it would predict the spray collection efficiency and choose the optimal surfactant from a database. The model underlying this algorithm would need to be robust with significant experimental data as input.

If a surfactant selection algorithm were developed through additional research, mine operators would have a tool to help them select the best surfactant spray for reducing respirable dust concentrations in their mines, ultimately leading to better respiratory health for miners.

Outcomes/Impact

Potential outcome: This project demonstrated that if it can be shown that respirable particles in a mine carry high levels of charge of a particular sign, then a surfactant carrying the opposite sign of charge should be chosen for the spray systems in that mine.

Potential outcome: This project demonstrated that if it can be shown that respirable particles in a mine are primarily neutral on average or carry only weak charges, then a non-ionic surfactant should be chosen for the spray systems in that mine.

Potential outcome: The findings from this project indicated a firm direction for future research. Further study is needed to understand the relationship between charge and size of surfactant spray drops and charge and size of respirable particles.

SECTION 2

Scientific Report

A. Background

Respirable dust exposure is a major health concern for miners in underground coal mines. Epidemiological studies have indicated that long term respirable dust exposure is associated with the development of coal workers' pneumoconiosis (CWP) and progressive massive fibrosis, causing a decrease in lung function and an increase in mortality even if the exposure level is at the current Mine Safety and Health Administration (MSHA) permissible exposure limit (PEL) of 2 mg/m^3 .⁽¹⁻⁴⁾ Even though the current PEL may not sufficiently protect miners from respirable coal dust exposure, some types of mining operations, especially longwall mining, still struggle to meet it. According to the National Institute for Occupational Safety and Health (NIOSH), 11% of mine operator samples for respirable dust were greater than 2.1 mg/m^3 during 2004–2008.⁽⁵⁾ Moreover, 25.6% of operator samples and 20.2% of inspector samples in underground coal mines were greater than NIOSH's recommended exposure limit (REL) of 1 mg/m^3 in 2003⁽⁶⁾, indicating that existing dust control technology in coal mines is not as effective as it could be in controlling respirable dust particles.

A variety of control measures have been adopted in underground coal mines to keep respirable dust levels below the current PEL, including reducing dust generation, preventing coal particles from becoming airborne, diluting dusty air with clean air, and capturing the airborne particles.⁽⁷⁾ In most underground coal mines, water-based sprays are a primary dust control technology owing to their superior cost-effectiveness compared to other options. Water-based sprays can be used both to wet the mining surface to prevent dust release and to capture those respirable particles that do become airborne.⁽⁸⁾ Although typical water-based sprays are effective at wetting mining surfaces, they are less efficient at capturing airborne respirable dust, with an average of 30% efficiency under actual mining conditions.⁽⁹⁾ Increasing spray collection efficiency may lower dust concentrations in mines and decrease the resulting health effects.

Surfactants are theorized to improve dust control mainly by increasing the wetting of the coal surface to prevent coal dust from becoming airborne.^(10, 11) The majority of coal dust particles generated during coal cutting stay attached to the coal surface.⁽¹²⁾ Because more dust stays attached to wetted coal surfaces than to dry surfaces,⁽⁸⁾ wetting the coal surface with a water spray during cutting is an important method for dust control. Many studies have documented the ability of surfactants to enhance the wetting of coal surfaces.⁽¹³⁻¹⁵⁾ In general they have found that surfactants, which have a hydrophilic head and hydrophobic tail (Figure 1), can

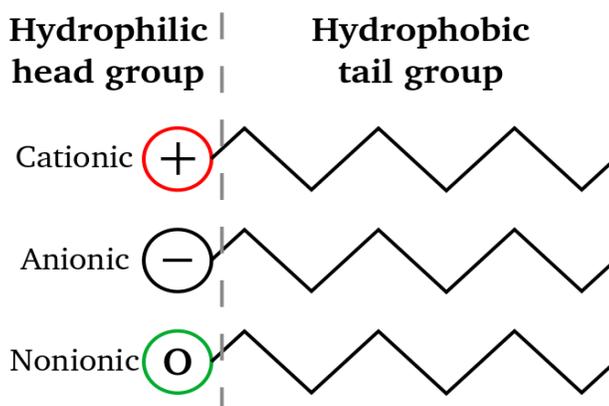


FIGURE 1. Surfactant classification based on charge present in its hydrophilic head.

change the interfacial tension between spray droplets and coal when the hydrophobic surfactant tail adsorbs to the hydrophobic coal surface, effectively changing the hydrophobic coal surface to a hydrophilic one⁽¹⁶⁾. In addition to enhancing the wettability, surfactants also decrease the surface tension of spray droplets, thereby decreasing the barrier for particles to enter the droplets⁽¹¹⁾ and effectively increasing the capture of airborne coal dust particles.

Although previous studies have shown that surfactants increase coal wettability, studies in coal mines have not come to a consensus about whether surfactants can improve spray collection efficiency. The improvement in respirable dust control efficiency while using surfactant-containing sprays compared to plain water has ranged from zero⁽¹⁷⁾ to 25%-30%⁽¹⁴⁾ to more than 40%⁽¹⁸⁾, and

even up to 60%⁽¹⁹⁾ in different studies. These varying results in different coal mines suggest that surfactant effectiveness may depend on some combination of the type of surfactant, coal type, dust size, and dust load.^(11, 20)

The surfactant effectiveness studies mentioned above, conducted in the 1980s and 1990s, only considered the hydrophobic nature of the coal and surfactant. Electrical attraction or repulsion between the charged coal dust particles and spray droplets were not considered. These electrical effects may impact spray efficiency,

causing the variability in effectiveness reported in previous studies. Although Walker et al.⁽²⁰⁾ pointed out the potential of using electrical attraction of dust and surfactant-containing spray droplets to enhance dust abatement, studies based on electrical properties of coal dust and surfactant were not available until the last decade.

Later studies on coal dust have observed that higher quality coals, with lower moisture content, exhibit a smaller fraction of dust in the respirable range. This is probably due to the stronger charging associated with the dust generated when these coals are pulverized. Stronger charging can lead to increased rates of particle agglomeration,^(21, 22) suggesting that coal dust particles could carry varying electrical charges depending on the characteristics of individual coal mines. In addition, surfactant studies have shown that spray droplets carry large numbers of elemental charges when they are generated.^(23, 24) Furthermore, the sign and the amount of charge varies by the classification (Figure 1) and concentration of surfactant used, indicating that anionic surfactant sprays tend to have a net negative charge, cationic sprays have a net positive charge, and nonionic surfactant sprays tend to have a small net positive charge.⁽²⁴⁾

B. Specific Aims

The long-term goal of this line of research is to reduce respirable dust concentrations in coal mines by improving engineering control technology. The electrical properties of coal dust and spray droplets suggest that adding the right type and concentration of surfactant to a water spray can produce predictably charged water droplets, allowing the use of electrostatic attraction between coal dust particles and surfactant-containing spray droplets with opposite polarities to enhance the capture of respirable coal dust.⁽²²⁾ The objective of this laboratory-based investigation was to measure how efficiently sprays containing different types and concentrations of surfactants collect particles having various levels of electrical charge. The objective was achieved by accomplishing the following three specific aims proposed in the original grant application:

1. Design and build a test apparatus to measure the ability of sprays to collect airborne particles,
2. Measure the spray collection efficiency for polystyrene latex (PSL) spheres using several spray surfactants, and
3. Measure the spray collection efficiency for a polydisperse coal dust using several spray surfactants.

This report describes our activities over the entire four-year project period.

C. Methods

We created a test apparatus that allows us to make measurements of collection efficiency by sprays containing surfactants as a function of particle diameter and charge sign and magnitude. Built to connect to the filtered entry plenum and to the fan + filtered exhaust of an existing wind tunnel, the portion of the apparatus constructed specifically for this project consists of three main sections: (1) a particle generation section, where the test particles were aerosolized, (2) a spray section, where sprays were generated and particles passed through the spray, and (3) a particle measurement section, where the size and charge level of particles were measured. These sections, measuring 1.83 m long and 30.5 cm x 30.5 cm in cross-section, were built in two pieces by the University of Minnesota Physics Research Machine Shop. Individual elements of each section were either already present in the Principal Investigator's laboratory or were purchased or manufactured.

A schematic diagram of the apparatus is shown in Figure 2. Photographs of the apparatus are presented in Figure 3. Air was drawn through the wind tunnel by a fan at a velocity of 0.61 m/s. Air entered the apparatus through a large HEPA filter bank to remove background particles, and then into a large plenum. The air moved from the plenum through a reducing transition into the main portion of the wind tunnel. After leaving the main section of the tunnel, the air passed through an expansion to the fan, and then went through another HEPA filter bank before returning to the room.

In the particle generation section, we generated two different types of particles. To address Specific Aim #2, we generated PSL particles with nominal diameters of 0.6, 1.0 and 2.1 μm (5060A, 5100A and 5200A, Thermo Scientific, Waltham, MA). To address Specific Aim #3, we generated a low-volatility bituminous coal dust (Austin Black 325, Harwick Standard, Akron, OH).

The three sizes of PSL spheres were made into separate suspensions with a concentration of 0.4% by volume using deionized and filtered water, and aerosolized using a homemade research-grade DeVilbiss-type

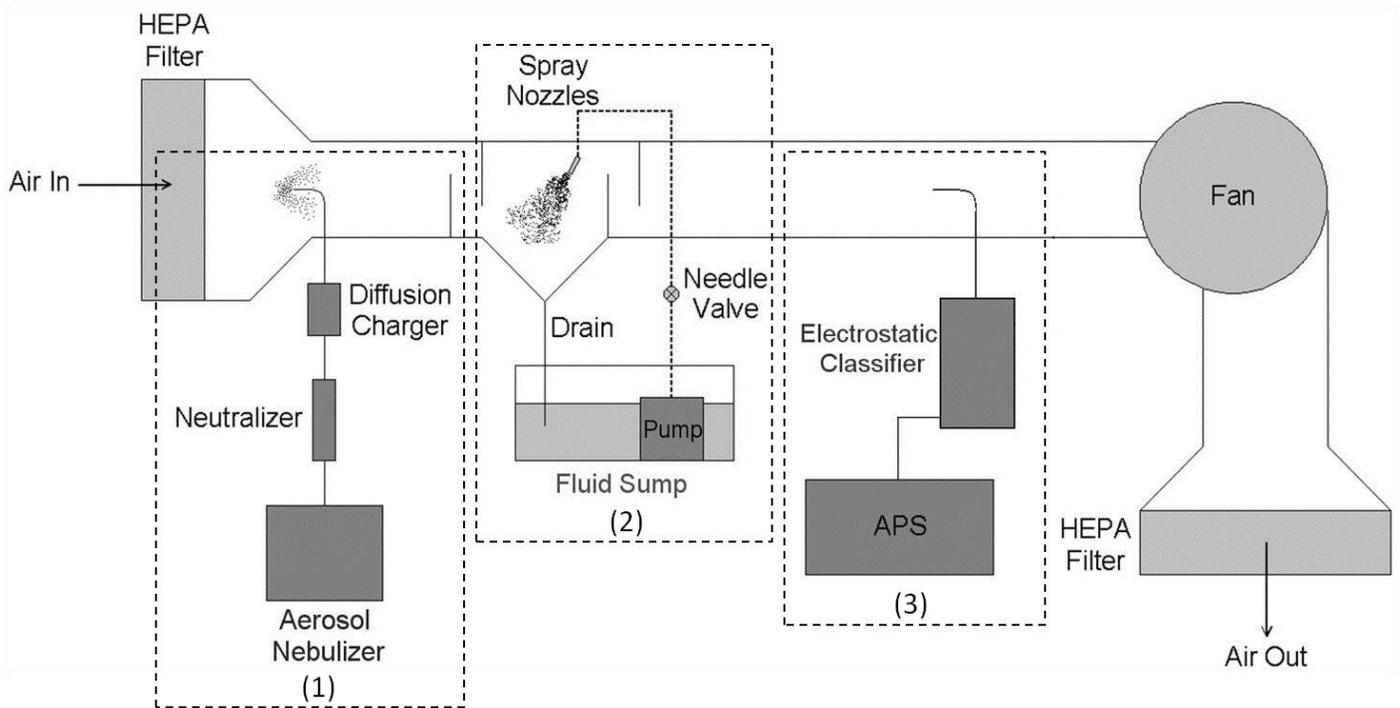
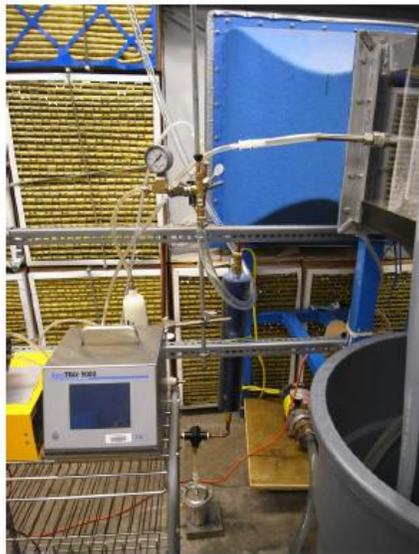


FIGURE 2. Schematic diagram of three apparatus sections: (1) the particle generation section, (2) the spray section, and (3) the particle measurement section, set in a wind tunnel.



Particle Generation Section



Spray Section



Particle Measurement Section

FIGURE 3. Images of the three primary sections of the test apparatus.

nebulizer at an air pressure of 40 psi. The water portion of the droplets formed during nebulization evaporated rapidly, leaving primarily individual spheres as test particles. This nebulizer was chosen because preliminary tests showed that it produced the most stable particle concentrations. As illustrated in Figure 4, preliminary tests also demonstrated that minimal airflow through the wind tunnel, high PSL concentration in nebulizer suspensions, and high air pressure to the nebulizer produced the highest particle concentrations. Furthermore,

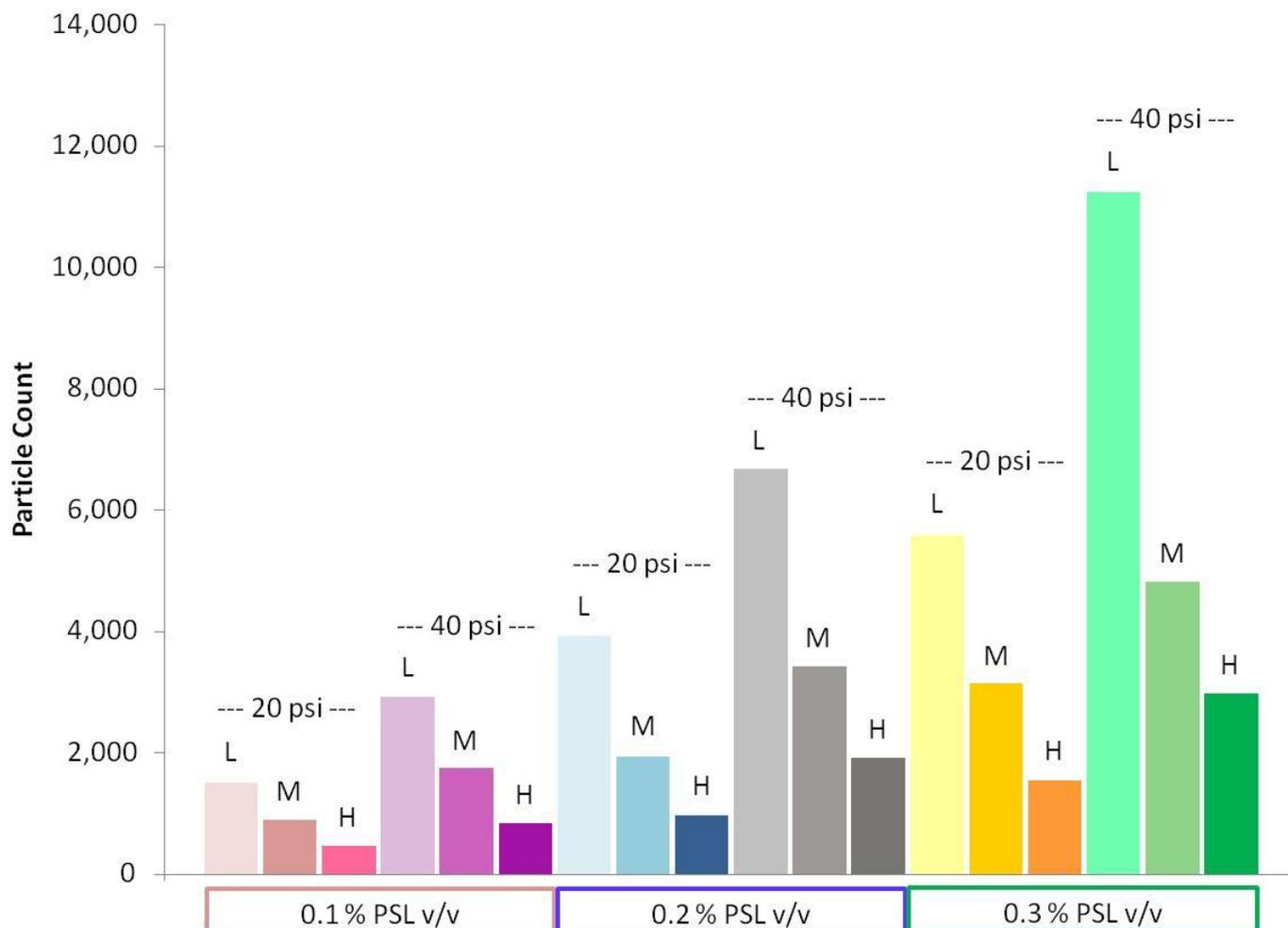


FIGURE 4. Particle counts generated in the test apparatus as a function of PSL concentration in the nebulizer (0.1%, 0.2%, or 0.3% v/v), nebulizer air pressure (20 or 40 psi), and wind tunnel air flow rate (L,M,H = low, medium, high).

the tests indicated that generated particle levels were steady throughout multi-hour tests, with a coefficient of variation of about 2%.

The coal dust was aerosolized using a Model 3400 Fluidized Bed Aerosol Generator (TSI Inc., Shoreview, MN). The dust was placed directly into the generator's reservoir. To produce particles that would be unneutralized or fully neutralized, the chain speed from the reservoir was set to 10-15 on the instrument and the bed flow was set to 50-55, equivalent to 12-13 L/min of air. For particles that would be imbued with a positive or negative charge, the chain speed was 30-50 and the bed flow was 7-10, equivalent to about 4.2-4.6 L/min. Chain speed and flow were adjusted regularly to obtain steady output over time.

After the particles were aerosolized, one of four aerosol charge conditions was established: (1) "unneutralized", where the aerosol produced by the nebulizer was injected into the wind tunnel without alteration to its charge, (2) "neutralized", where the aerosol passed through an Aerosol Neutralizer (Model 3012A, TSI Inc.) which used a Kr-85 beta particle radiation source to imbue the aerosol with a Boltzmann equilibrium charge distribution before the particles were injected into the wind tunnel, (3) "negative", where the aerosol passed first through the neutralizer and then through a unipolar diffusion charger taken from a Nanoparticle Aerosol Monitor (AeroTrak 9000, TSI Inc.) operating with a voltage of -5.3 kV supplied by an Eisco 5kV regulated DC power supply (Eisco Labs, Ambala Cantt, India), or (4) "positive", which was identical to negative charge condition but with the diffusion charger operating at a +5.3 kV voltage. In the charger, the particles received their charge as the aerosol flow mixed with ions generated by a high voltage applied corona

needle. The generated aerosol was then injected into the wind tunnel through an inlet pointed in the direction opposite to the air flow to maximize mixing.

After leaving the generation section, particles traveled with the air flow through the wind tunnel to the spray section. In this section, the spray solution was made using one of the following types of surfactant: anionic (Sodium dodecyl sulfate, Fisher Scientific Inc., NJ) at either 10^{-4} M ("low concentration") or 10^{-6} M ("high concentration"), nonionic (Triton X-100, Fisher Scientific Inc., NJ) at either low or high concentration, or cationic (Dodecylamine hydrochloride, Tokyo Chemical Industry Co., Japan) at either low or high concentration, or plain deionized water as a control. The spray solution was stored in a 190 L container and pumped to three hollow cone spray nozzles (No. TTD4-46, Spraying Systems Co., Wheaton, IL). These nozzles were among those used by Pollock and Organiscak⁽²⁵⁾ to evaluate the influence of nozzles on dust collection. The spray solution ran at a flow of 2.95 L/min to each nozzle at a pressure of 552 kPa, and then drained back into the sump and was recirculated by a pump (Model 4UP51A, Dayton Electric Manufacturing Co., Niles, IL) through a polypropylene filter (Model 08PT, Keystone Filter, Hatfield, PA) which prevented captured particles from being reinjected into the wind tunnel. Some particles were removed by the spray and the others passed through the spray section along with the air flow.

After leaving the spray section, the remaining particles moved through the wind tunnel, and some of them were sampled by a probe pointed into the flow. The probe was designed according to specifications by Brockmann et al.⁽²⁶⁾ that take into account both aspiration efficiency into the probe and transport efficiency through the probe for an optimal sampling velocity that was somewhat subisokinetic. This probe design method was employed successfully by Raynor et al.⁽²⁷⁾ in the same wind tunnel. Sampled particles were separated according to their electrical mobility by an electrostatic classifier (Model 3071A, TSI Inc.) which used an external voltage source (Bertran 230 high voltage power supply, Spellman High Voltage Electronics Corporation, Valhalla, NY) capable of providing both signs of charge. As the voltage was varied, different mobility ranges were sampled by the classifier. The particles that passed through the classifier were counted and sized in an Aerodynamic Particle Sizer (APS, Model 3321, TSI Inc.). The number of elementary charges on the particle, n , was calculated as

$$n = \frac{3Z_p \pi \mu D_p}{eC} \quad (1)$$

in which Z_p is electrical mobility, μ is gas viscosity, D_p is particle diameter, e is elementary charge, and C is Cunningham slip correction.⁽²⁸⁾

To measure collection efficiency, particles were counted both with and without the spray applied, as shown in Table I. The net particle count

when the spray was not applied, C_{SprayOff} , was obtained by subtracting the background particles in the wind tunnel from the total test particles generated. The net particle count when the spray was applied, C_{SprayOn} , was obtained by subtracting the count of net spray droplets from the total count of particles and background particles that were not removed by the spray. Collection efficiency, η , is then calculated as

$$\eta = 1 - \frac{C_{\text{SprayOn}}}{C_{\text{SprayOff}}} = 1 - \frac{(B_{\text{SprayOn}} + P_{\text{SprayOn}} + D) - (B_{\text{SprayOn}} + D)}{(B_{\text{SprayOff}} + P_{\text{SprayOff}}) - B_{\text{SprayOff}}} \quad (2)$$

Two kinds of collection efficiency were investigated. The overall collection efficiency, η_{overall} , for particles of all charge levels was measured when sampled particles were directly routed to the APS without passing through

TABLE I. Four measurements for efficiency calculations.

Test setup		Measurement	
Particle generation	Spray		
Off	Off	B_{SprayOff}	Background particles
On	Off	$B_{\text{SprayOff}} + P_{\text{SprayOff}}$	Background particles and test particles combined
On	On	$B_{\text{SprayOn}} + P_{\text{SprayOn}} + D$	Background particles and test particles that are not removed by spray, plus spray droplets
Off	On	$B_{\text{SprayOn}} + D$	Background particles that are not removed by spray, plus spray droplets

the electrostatic classifier. Using the APS, particles were counted in narrow size intervals so that efficiency could be differentiated as a function of particle diameter. In addition, collection efficiencies for particles with specific charge levels, η_{charge} , were investigated by sampling the particles with the classifier at specific voltages and then counting them with the APS.

The influence on η_{overall} of particle diameter, aerosol charge condition, and surfactant classification and concentration was measured using PSL particles. Controlled conditions for each independent variable are given in Table II. The combination of 3 particle diameters, 4 charge conditions, 3 surfactants, and 2 surfactant concentrations yielded a total of 72 test conditions. Plain water spray with 3 particle diameters and 4 charge conditions added 12 additional conditions, for a total of 84 test conditions. Three separate replicate tests were conducted for each combination of conditions. In addition, η_{charge} was measured for 0.6 and 1.0 μm diameter PSL particles at 8 specific voltages. The influence on η_{charge} of aerosol charge condition and surfactant classification and concentration was also evaluated for a total of 2 diameters x 4 charge conditions x 7 surfactants x 8 voltages = 448 combinations of test conditions, each measured three separate times.

For the coal dust, η_{overall} was measured using the default size intervals on the Aerodynamic Particle Sizer for particles ranging from about 0.54 μm in aerodynamic diameter to about

3.3 μm . The upper limit varied from test to test because few large particles were able to pass through the charging devices. Tests were conducted in triplicate for each combination of the four aerosol charge conditions and the high concentrations of the three surfactants listed in Table II plus plain water. This resulted in a total of 48 tests (4 charging conditions x 4 spray fluids x 3 replicates).

The effects of particle diameter, aerosol charge condition, surfactant classification, and surfactant concentration on overall collection efficiency were determined statistically using a multi-factor analysis of variance (ANOVA). The effects of particle charge level, surfactant classification and surfactant concentration on the collection efficiency of particles with specific charge levels were also determined statistically using a multi-factor ANOVA. Scheffe's method was used to evaluate the effects of aerosol charge condition, surfactant classification, and spray solution type on the arithmetic means of collection efficiency. Spray solution type is defined as a specific combination of surfactant classification and concentration, including plain water. The plots of arithmetic mean and standard deviation of collection efficiencies were calculated based on three replicates of experiments. Statistical analyses were performed using R project version 2.13.0.⁽²⁹⁾ Overall coal dust efficiency as a function of particle diameter was compared graphically to the overall efficiency of the PSL particles to determine if the efficiencies for the two kinds of particles are comparable.

D. Results

Overall Collection Efficiency with PSL Particles

Overall collection efficiency using the PSL particles was significantly affected by particle diameter and spray solution type ($p < 0.0001$ and $p = 0.033$ respectively), but not by aerosol charge condition ($p = 0.76$). Particle diameter had the largest impact on spray overall collection efficiency ($p < 0.0001$), as shown in Figure 5. The overall collection efficiency was $21.5\% \pm 9.0\%$ (mean \pm SD) for 0.6 μm PSL particles, $58.8\% \pm 12.5\%$ for 1.0 μm particles, and $86.6\% \pm 43.5\%$ for 2.1 μm particles across all types of surfactant-containing sprays regardless

TABLE II. Test Conditions for Efficiency Measurements

Independent Factors	Test Condition
Particle Diameter	0.6 μm
	1.0 μm
	2.1 μm
Aerosol Charge Condition	Unneutralized
	Neutralized
	Negative
	Positive
Surfactant Classification	Anionic (Sodium lauryl sulfate)
	Nonionic (Triton X-100)
	Cationic (Dodecylamine hydrochloride)
Surfactant Concentration	0 (Plain water)
	1×10^{-6} M (Low concentration)
	1×10^{-4} M (High concentration)

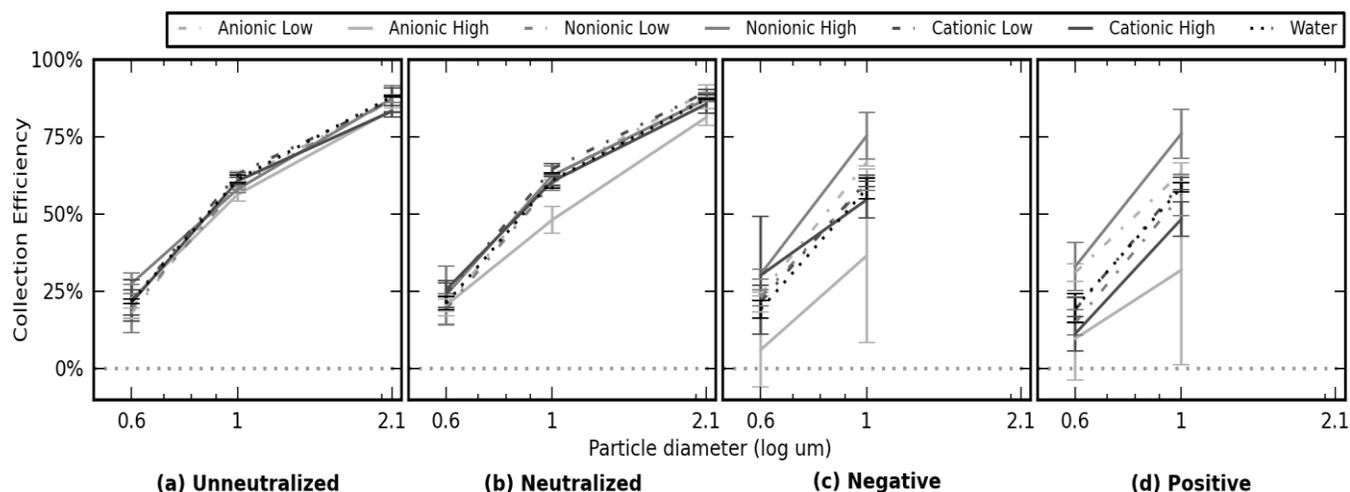


FIGURE 5. Overall collection efficiency (mean±SD) as a function of particle diameter for four particle charge conditions.

of aerosol charge condition, suggesting that effect of aerosol charge condition, surfactant classification, and surfactant concentration on overall collection efficiency may be different for each particle diameter.

Aerosol charge condition did not significantly affect the overall collection efficiency. There were no significant differences among the four tested aerosol charge conditions for the 0.6, 1.0 or 2.1 μm particle diameters. P-values were $p=0.78$, $p=0.67$, and $p=0.53$, respectively.

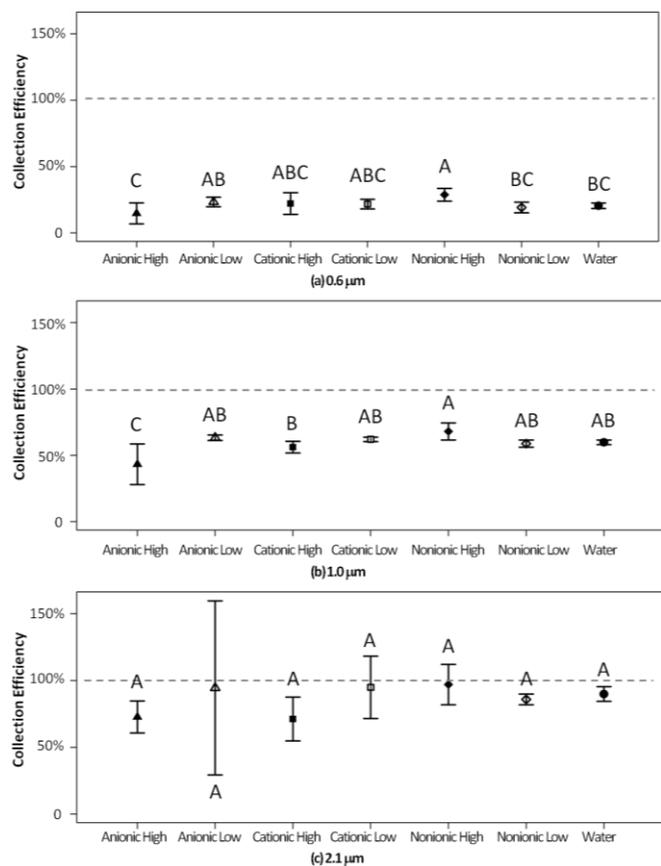


FIGURE 6. Mean overall collection efficiency with 95% confidence intervals as a function of spray solution type for three particle diameters. Means with different letters are significantly different (Scheffe's test, $p<0.05$).

Spray solution type significantly affected the overall collection efficiency for both 0.6 and 1.0 μm particles ($p=0.0087$ and $p<0.0001$ respectively) regardless of the aerosol charge condition (Figure 6), suggesting that the high concentration nonionic surfactant-containing spray had the best performance capturing 0.6 and 1.0 μm particles regardless of the aerosol charge condition, while the high concentration anionic surfactant-containing spray had the lowest particle suppression. The lack of significance observed for 2.1 μm particles ($p=0.65$) was related to the larger variance in collection efficiency for 2.1 μm particles which masked any differences among the seven spray solution types.

Although the effect of surfactant classification on overall spray collection efficiency was not statistically significant ($p=0.43$), the nonionic surfactant-containing spray consistently had the highest collection efficiency for all three particle diameters, as shown in Table III. Nonionic surfactant-containing sprays had the highest collection efficiency for both negatively- and positively-charged aerosols. The cationic surfactant-containing sprays had relatively high efficiency for negatively-charged aerosols and the lowest efficiency for positively-charged aerosols, while the anionic surfactant-containing sprays had relatively high efficiency for positively-charged aerosols and the lowest efficiency for negatively-charged aerosols. All three surfactant-containing

TABLE III. Comparison of mean overall collection efficiency by surfactant classification and particle diameter.

Surfactant Classification	Diameter = 0.6 μm			Diameter = 1.0 μm			Diameter = 2.1 μm		
	N	Mean	SD	N	Mean	SD	N	Mean	SD
Anionic	24	19.1%	10.4%	24	53.3%	19.6%	24	83.6%	72.9%
Nonionic	24	24.0%	8.4%	24	63.4%	8.9%	24	91.5%	17.9%
Cationic	24	22.0%	9.7%	24	59.1%	5.9%	24	83.1%	33.3%
Water	12	20.5%	3.3%	12	59.8%	2.8%	12	89.9%	8.6%

sprays and the plain water spray had similar efficiency for neutralized and unneutralized aerosols. Results for 0.6 μm particles are shown in Figure 7, and the results for 1.0 and 2.1 μm particles exhibit similar patterns.

Surfactant concentration significantly affects the overall collection efficiency for anionic and nonionic surfactant-containing sprays on 0.6 and 1.0 μm particles, and for cationic surfactant-containing sprays on 1.0 μm particles, as shown in Table IV. There was no difference in overall collection efficiency for 2.1 μm particles among the three surfactant concentration levels for anionic, nonionic and cationic surfactants owing to the large variance in the results. Although spray solution type significantly influenced overall spray collection efficiency, surfactant concentration did not affect spray collection consistently across surfactant classifications.

Collection Efficiency for PSL Particles at Specific Charge Levels

Figure 8 shows the particle collection for three classifications of high concentration surfactant-containing sprays for particles having various charge levels at the four aerosol charge conditions. Surfactant classification had a significant impact on spray collection efficiency for most charge levels. In addition, it is apparent that collection efficiency is a function of aerosol charge, even though aerosol charge condition did not have a significant effect on overall collection efficiency.

Unneutralized aerosols tended to have more highly charged particles with both signs, neutralized aerosols tended to have fewer charged particles and those particles tended to have fewer charges, and negative and positive aerosols tended to have more charged particles with only negative or positive signs. Because different charge conditions cause differing charge distributions, using different surfactant-containing sprays causes differing reactions among charge conditions. For neutralized aerosols, the high concentration anionic surfactant-containing spray significantly removed most positively-charged particles, but not negatively-charged particles. Conversely, the high concentration cationic surfactant-containing spray significantly removed most negatively-charged particles, but not positively-charged particles. The high concentration nonionic surfactant-containing spray significantly removed both negatively- and positively-charged particles, but not as efficiently as the ionic surfactants for oppositely-

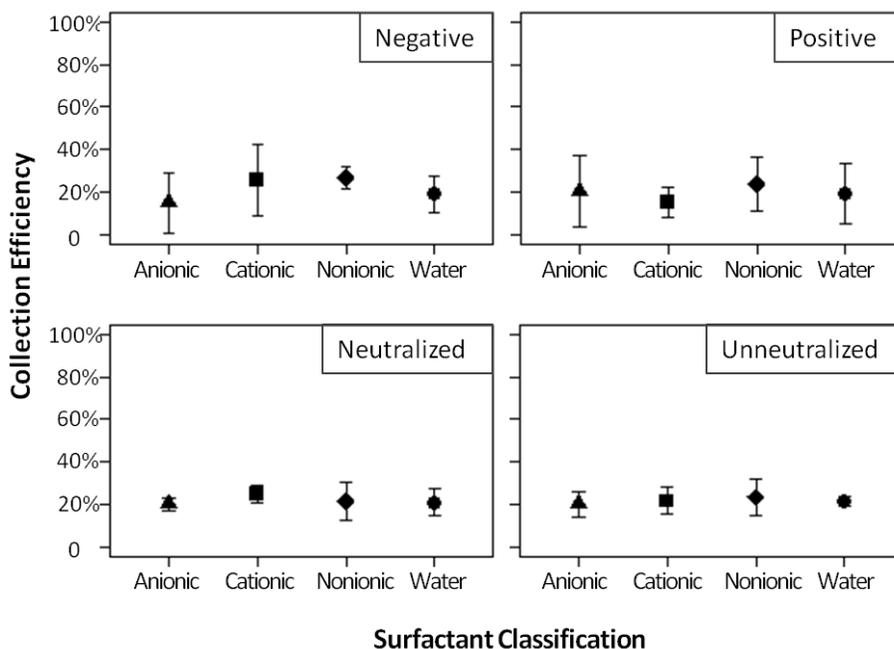


FIGURE 7. Mean overall collection efficiency with 95% confidence intervals as a function of surfactant classification for 0.6 μm particle with the four aerosol charge conditions.

TABLE IV. Comparison of mean overall collection efficiency by surfactant classification, concentration and particle diameter. Twelve measurements were averaged for each combination of classification, concentration, and diameter. Bold p-values are significant at $p < 0.05$.

Classification	Conc.	Diameter = 0.6 μm			Diameter = 1.0 μm			Diameter = 2.1 μm		
		Mean	SD	p-value	Mean	SD	p-value	Mean	SD	p-value
Anionic	0	20.5%	3.3%		59.8%	2.8%		89.9%	8.6%	
	Low	23.4%	5.7%	0.014	63.3%	3.2%	0.0047	94.5%	102.5%	0.64
	High	14.8%	12.4%		43.3%	24.0%		72.8%	18.8%	
Nonionic	0	20.5%	3.3%		59.8%	2.8%		89.9%	8.6%	
	Low	19.2%	6.3%	0.00038	58.8%	4.2%	0.00039	85.9%	6.3%	0.18
	High	28.8%	7.5%		68.0%	10.2%		97.0%	23.8%	
Cationic	0	20.5%	3.3%		59.8%	2.8%		89.9%	8.6%	
	Low	21.8%	5.7%	0.87	62.1%	2.6%	0.0023	94.9%	36.8%	0.11
	High	22.2%	12.9%		56.1%	6.9%		71.2%	25.8%	

charged particles. For the unneutralized and positively- or negatively-charged aerosols, both ionic surfactant-containing sprays showed better performance on capturing particles with opposite charge than on capturing particles with same charge. However, nonionic surfactant-containing spray showed superior performance on capturing highly charged particles with both signs of charge.

Particle charge level also impacted spray collection efficiency for both 0.6 and 1.0 μm particles ($p < 0.0001$). A significant increase in spray collection efficiency was observed with increased particle charge for both 0.6 and 1.0 μm particles ($p < 0.0001$), as shown in Figures 9 and 10. The average collection efficiency for 0.6 μm particles decreased from $48.1\% \pm 22.2\%$ (mean \pm SD) for 56 charges per particle to $-38.7\% \pm 37.6\%$ for 9 charges across the seven sprays, and average collection efficiency for 1.0 μm particles decreased from $72.8\% \pm 10.1\%$ for 102 charges per particle to $32.4\% \pm 26.5\%$ for 17 charges.

Within each particle charge level, spray solution type had a substantial effect on collection efficiency. Surfactant classification contributed to this effect more than surfactant concentration, especially for 0.6 μm particles. As shown in Figures 9 and 10, Scheffe's method indicated that cationic surfactant-containing sprays had significantly higher collection

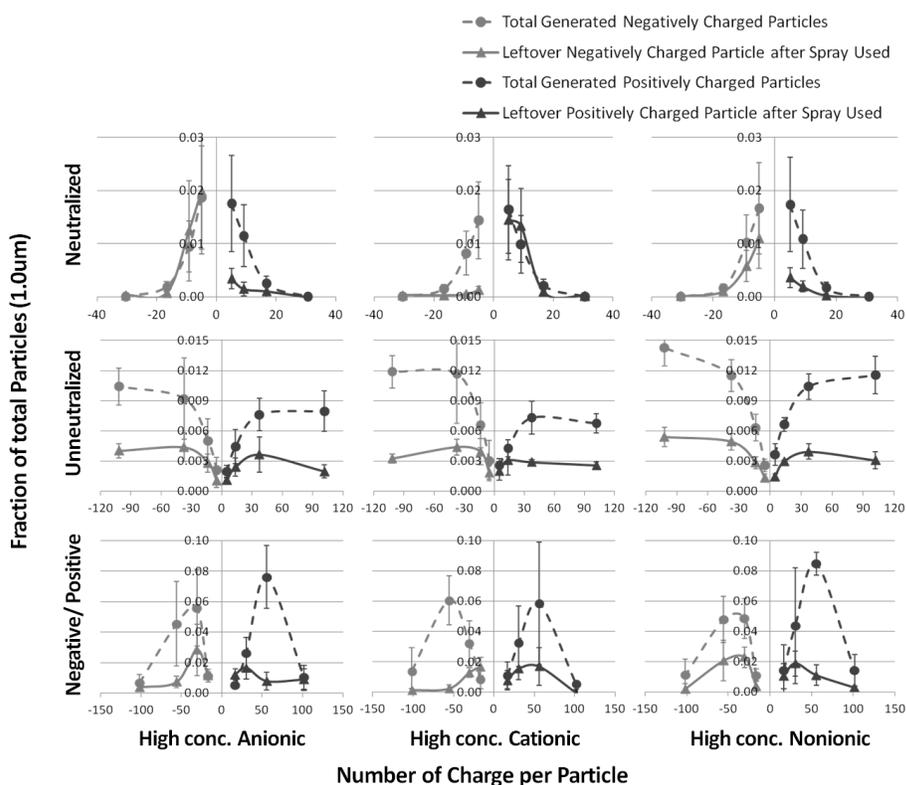


FIGURE 8. Fraction of total PSL particles (mean \pm SD) generated with spray off or on by three surfactant sprays for 1.0 μm particles for each aerosol charge condition. Neutralized and unneutralized particles are shown in the first two rows. Negatively (left) and positively (right) charged particles were combined in the third row.

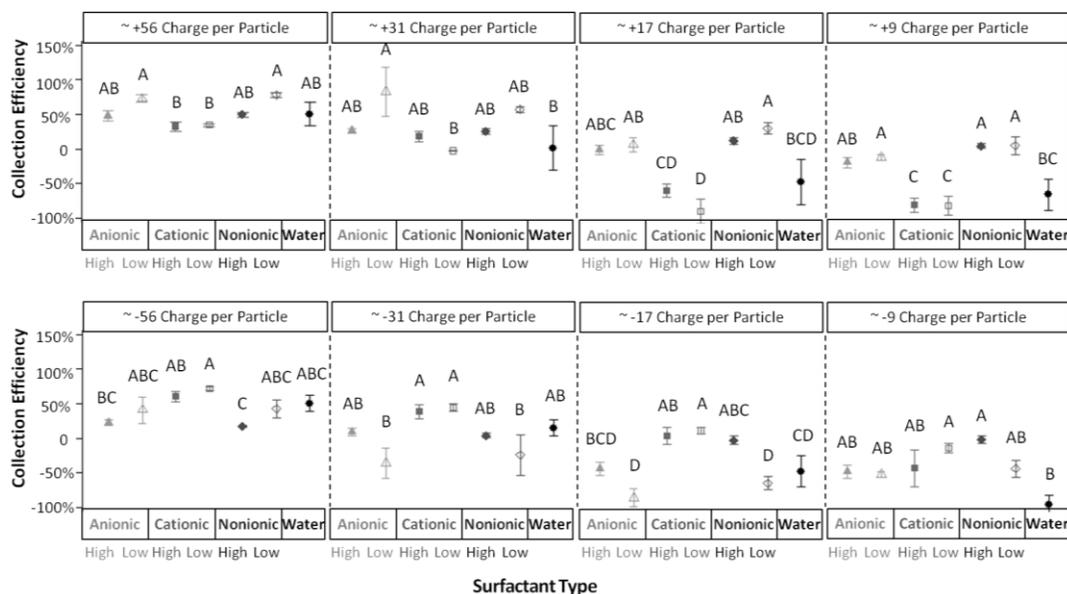


FIGURE 9. Collection efficiency (mean±SD) as a function of spray solution type for different charge levels of 0.6 μm particles. Means with different letters are significantly different (Scheffe's test, $p<0.05$).

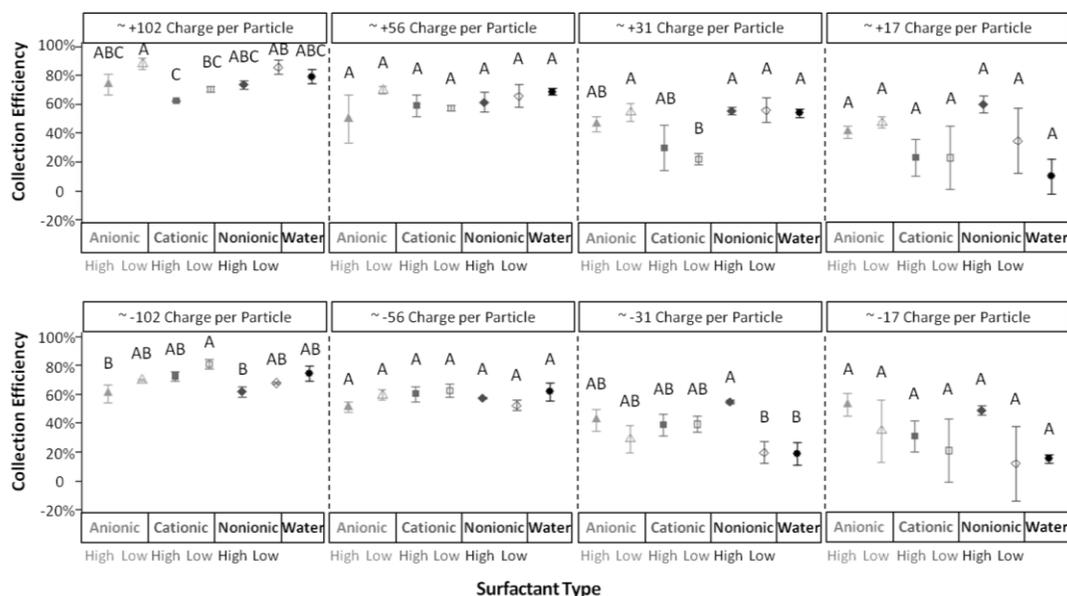


FIGURE 10. Collection efficiency (mean±SD) as a function of spray solution type for different charge levels of 1.0 μm particles. Means with different letters are significantly different (Scheffe's test, $p<0.05$).

surfactant, including water, and aerosol charge condition. More error was observed for the negatively- and positively-charged particles because passing through the diffusion charger reduced particle concentrations. Agreement between coal dust particles and the PSL particles was excellent for unneutralized and neutralized particles. The negatively- and positively-charged particles exhibited poorer agreement, especially for the 2.1 μm PSL particles. Nonetheless, the agreement was still reasonable even for these charge conditions.

efficiencies for negatively-charged particles, and significantly lower collection efficiencies for positively-charged particles, compared to other sprays. This difference was largest for highly charged particles. Conversely, anionic surfactant-containing sprays had significantly higher collection efficiencies for positively-charged particles and significantly lower collection efficiencies for negatively-charged particles, compared to other sprays. Nonionic surfactant-containing sprays had inconsistent collection efficiency for charged particles. The collection efficiency of water spray for charged particles was between the two ionic surfactant-containing sprays.

Overall Collection Efficiency with Coal Dust Particles

Figure 11 shows comparisons between spray collection efficiency for the 0.6, 1.0, and 2.1 μm PSL particles and efficiency for the coal dust across all diameter intervals from 0.54 to 3.3 μm . Comparisons are made for all combinations of

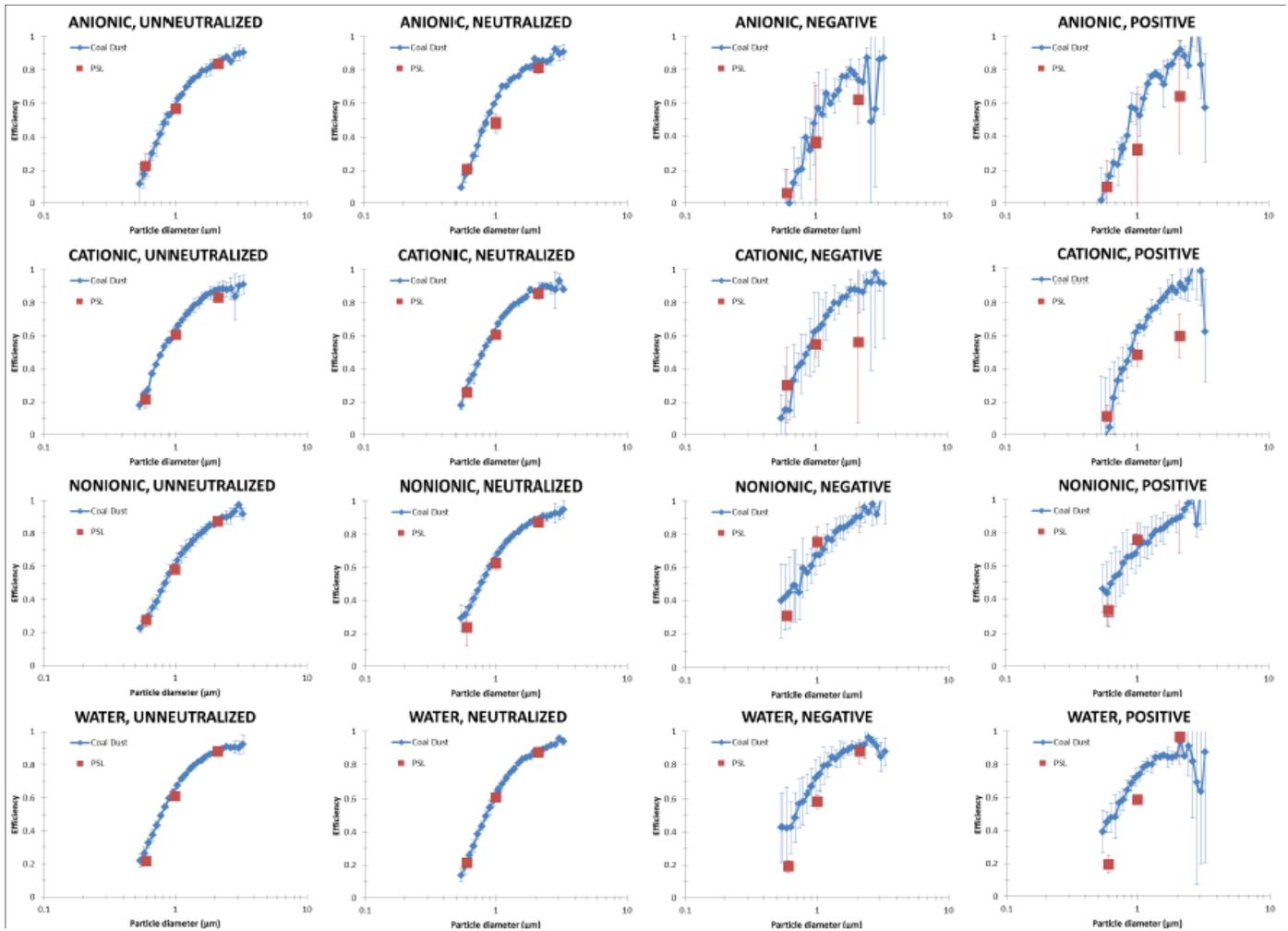


FIGURE 11. Collection efficiency for unneutralized, neutralized, negatively-charged, and positively-charged PSL particles and coal dust particles captured by anionic, cationic, nonionic, and water sprays. Comparisons between the two types of particles are made for each combination of charge condition and spray. Data points and error bars represent the mean and standard deviation for three separate measurements.

E. Discussion

The capture of aerosol by water spray involves the physical mechanisms of inertial impaction, interception, gravity, diffusion, and electrical effects. This study focused on respirable particles with diameters ranging from 0.6 to 2.1 μm and charges ranging from 0 to 120 charges per particle. Because particles with this diameter range were mainly removed by inertial impaction,⁽³⁰⁾ particle diameter was the dominant factor impacting both the overall spray collection efficiency and the collection efficiency for particles with different charge levels. Significantly higher capture efficiencies were observed among larger particles (Figure 5) regardless of test particle charge levels (Figures 9 and 10), implying that particle capture was less impacted by electrical effects than inertial impaction.

In this study, an average of 86.6% of 2.1 μm particles were removed by the spray, indicating that coal mines with a greater proportion of coarse dust particles ($>2 \mu\text{m}$) are likely have more effective dust control by spray and less room for improvement. Therefore, coal mines with finer dust particles ($<2 \mu\text{m}$) may be the best targets for improvements in spray collection efficiency.

Aerosol charge condition, surfactant classification, and surfactant concentration had no significant effect on overall collection efficiency, indicating that the various improvements on respirable dust control found in previous studies^(14,17-19) using surfactant-containing sprays may have been caused by other surfactant

properties, such as surface-tension-lowering ability and polarity, rather than electrical characteristics. The spray solution type had significant effect on overall collection efficiency across aerosol charge conditions, indicating that the combination of surfactant classification and concentration is important for respirable dust control. The high concentration nonionic surfactant-containing spray suppressed at least 5% more 0.6 and 1.0 μm diameter particles than other surfactant solutions and at least 8% more than plain water as shown in Table IV. Similar findings have been reported by Tien and Kim, ⁽¹¹⁾ and they attribute the efficacy of the nonionic surfactant to its superior coal wetting qualities. The difference in overall collection efficiency among the tested surfactants was not statistically significant for aerosols with particle charges ranging from 0 to 120 charges. However, the differences in overall collection efficiency may be more significant for coal dust particles, which may carry more surface charges than the average charges of the particles tested in this study. According to a survey in U.S and French mines, coal dust particles with a diameter of 60 to 90 μm carried up to 1×10^{10} charges per particle.⁽³¹⁾ Respirable coal dust particles in mines will carry far fewer charges than these large particles do, but they are likely to be more highly charged than the particles in our study. Therefore, it is useful to investigate how particle charge level impacts overall collection efficiency.

Compared to the overall collection efficiency, which reflects the collection of particles carrying a range of charges, the collection efficiency for particles at specific charge levels reveals how different signs and magnitudes of particle charge influence particle capture. Analysis at individual charge levels shows that the aerosol charge condition, which did not significantly affect overall collection efficiency, was more important for collection efficiency for particles at specific charge levels. The reason for this discrepancy is that the various charge conditions give aerosol charge distributions with some particle charge levels that are efficiently removed and some that are not.

Since the charge-specific collection efficiency can explain the electrical effects between charged particles and surfactant-specific sprays, it is a relevant property for surfactant selection. For negatively-charged aerosols, cationic and nonionic surfactant-containing sprays had higher overall collection efficiencies; for positively-charged aerosols, anionic and nonionic surfactant-containing sprays had slightly higher efficiencies than other sprays (Figure 7). Although there is no statistical difference among those spray solutions, if the average charge of the dust in a coal mine is known, using an ionic surfactant-containing spray with the opposite charge may yield optimal dust capture. However, if the averaged charge properties of the dust have not been quantified, results suggest that it may be worth considering a non-ionic surfactant like Triton X-100.

Certain aerosol charge conditions are more sensitive to surfactant use than others. As shown in Figure 8, neutralized and positively- and negatively-charged aerosols are more sensitive to surfactant solution use. However, unneutralized aerosols can be efficiently removed by any surfactant-containing spray, with the best removal efficiency by nonionic surfactant-containing spray. This observation implies that if coal dust aerosol has a weak or highly unipolar charge, the collection efficiency may be more sensitive to surfactant selection. Particle charge level also impacted spray collection efficiency in that highly charged particles tended to be removed more efficiently than weakly charged particles for both 0.6 and 1.0 μm particles (Figures 9 and 10). Although ionic surfactant-containing sprays also captured more particles with the opposite charge than with the same charge, the differences were not always significant, indicating that highly charged particles tended to be removed easily regardless of spray solution type. Finally, spray solution type had a significant and consistent effect on dust capture for aerosols with a known charge distribution. For highly unipolar-charged particles, the surfactant-containing sprays with the opposite charge were always significantly superior to the surfactant-containing sprays with the same charge and to water spray for dust suppression (Figures 9 and 10). For weakly bipolar charged aerosols, ionic surfactant-containing sprays significantly removed particles with the opposite charge but did not efficiently remove particles with same charge, as shown in Figure 8, while nonionic surfactant-containing sprays significantly removed aerosols with both signs of charge, with more efficient removal for positively-charged particles.

An additional finding in this study is that negative collection efficiencies as calculated using equation (2) were seen among weakly charged 0.6 μm particles (Figure 9). One possible explanation for this phenomenon would be that, as the PSL particles carried more charges per particle than the spray droplets, when spray droplets and PSL particles collided, there may have been a transfer of electric charge from the highly charged PSL particles to the weakly charged or nearly neutral spray droplets. After those droplets gained charges, they moved through the wind tunnel with the air flow, decreased in size due to evaporation, and then were measured by the APS. This introduction of additional weakly charged spray droplets into the system could

cause the measurement of $B_{\text{Spray On}} + P_{\text{Spray On}} + D$ to increase, which would decrease the calculated collection efficiency. This charge transfer would happen consistently among all experiments, and thus should not affect the assessment of relative collection efficiencies among the various test conditions. The further understanding and testing of this hypothesis is an area for future research.

In general, the optimal surfactant for an application varies based on the electrical properties of the coal dust to be removed. If a coal dust aerosol contains mainly highly charged unipolar particles, using an ionic surfactant-containing spray with the opposite charge could improve the dust capture compared to using a water spray. However, if a coal dust aerosol contains mainly weakly charged particles, using a high concentration nonionic surfactant-containing spray could yield the best respirable dust capture. Information on the charge distribution of coal dust is important for choosing the optimal surfactant, but if this information is not available, the average charge of coal dust could also be helpful.

As shown in Figure 11, spray collection efficiency for coal dust particles is approximately the same as the efficiency for PSL particles for similar particle sizes. Thus, PSL particles are a reasonable surrogate for coal dust in these measurements. Although the differences in efficiency are more pronounced for the negatively-charged and positively-charged particles, the error bars are considerably larger. Therefore, many of the observed differences are not significant statistically.

F. Conclusions

Particle size is the most important determinant of the spray collection efficiency for respirable particles. Dust particles with a diameter larger than $2 \mu\text{m}$ can be effectively removed by spray. Therefore, a choice of surfactant to improve dust collection by electrostatic attraction will primarily be important for smaller particles. Surfactant type significantly impacts collection of respirable dust by aqueous sprays. Adding ionic surfactant into spray water can improve dust capture for particles with a highly opposite charge. High concentration nonionic surfactant-containing spray can improve dust capture for particles with weak charge. Particle charge level impacts spray collection efficiency. Highly charged particles can be removed more efficiently than weakly charged particles. Therefore, highly charged dust is better collected by spray than weakly charged dust, especially by the spray droplets with opposite charge. Knowledge of the charge distribution of an aerosol, or at least the average sign and magnitude of charge on an aerosol, is important for informing surfactant choice.

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Publications

The first manuscript from the project has been reviewed for publication in *Journal of Occupational and Environmental Hygiene* and was returned to us on December 10, 2013 for minor revisions. Thus, we expect the manuscript will be revised, accepted, and in press soon. A citation for this manuscript is:

Principal Investigator: Raynor, Peter C.

Mei W. Tessum, Peter C. Raynor, and Lorraine Keating-Klika, "Factors Influencing the Control of Respirable Charged Particles by Surfactant Sprays ", submitted to *Journal of Occupational and Environmental Hygiene*.

We expect to submit a second manuscript on the coal dust particle experiments to another research journal within the next six months.

We have made presentations on this research at the American Industrial Hygiene Conference and Exposition (AIHce) in 2012 in Indianapolis and 2013 in Montreal.