



Contents lists available at ScienceDirect

Safety and Health at Work

journal homepage: www.e-shaw.org

Original Article

Effects of Spray Surfactant and Particle Charge on Respirable Coal Dust Capture



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ARTICLE INFO

Article history:

Received 22 May 2015

Received in revised form

25 October 2016

Accepted 12 December 2016

Available online 6 February 2017

Keywords:

efficiency

electric charges

respirable coal dust

spray

surfactant

ABSTRACT

Background: Surfactant-containing water sprays are commonly used in coal mines to collect dust. This study investigates the dust collection performance of different surfactant types for a range of coal dust particle sizes and charges.

Methods: Bituminous coal dust aerosol was generated in a wind tunnel. The charge of the aerosol was either left unaltered, charge-neutralized with a neutralizer, or positively- or negatively-charged using a diffusion charger after the particles were neutralized. An anionic, cationic, or nonionic surfactant spray or a plain water spray was used to remove the particles from the air flow. Some particles were captured while passing through spray section, whereas remaining particles were charge-separated using an electrostatic classifier. Particle size and concentration of the charge-separated particles were measured using an aerodynamic particle sizer. Measurements were made with the spray on and off to calculate overall collection efficiencies (integrated across all charge levels) and efficiencies of particles with specific charge levels.

Results: The diameter of the tested coal dust aerosol was $0.89 \mu\text{m} \pm 1.45$ [geometric mean \pm geometric standard deviations (SD)]. Respirable particle mass was collected with $75.5 \pm 5.9\%$ (mean \pm SD) efficiency overall. Collection efficiency was correlated with particle size. Surfactant type significantly impacted collection efficiency: charged particle collection by nonionic surfactant sprays was greater than or equal to collection by other sprays, especially for weakly-charged aerosols. Particle charge strength was significantly correlated with collection efficiency.

Conclusion: Surfactant type affects charged particle spray collection efficiency. Nonionic surfactant sprays performed well in coal dust capture in many of the tested conditions.

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1. Introduction

Coal worker's pneumoconiosis, which is caused by prolonged exposure of airborne respirable coal dust, is one of the major occupational diseases affecting coal miners. Although great efforts have been taken in underground coal mines to control respirable coal dust, the exposure level of respirable coal dust still often exceeds occupational exposure limits [1].

Water spray systems are one of the widely used control techniques for prevention and suppression of respirable coal dust in coal mines. Since the 1960s, surfactants have been suggested as additives to spray water with the intention of preventing airborne dust release and improving the efficiency of dust control based on

the theory that surfactants can enhance wettability, reduce the surface tension of water, and allow dust particles to better penetrate the water drops [2–5]. Although surfactant sprays are widely used, their ability to enhance the capture of airborne respirable coal dust varies from mine to mine [6,7]. Some studies have attributed these differences mostly to factors such as surface tension, wettability, and water mineralogy that influence penetration and adsorption of coal dust particles into spray drops [4,5]. Other studies have hypothesized that electrical attraction or repulsion between dust particles and spray drops also impacts dust collection [8].

Previous studies have provided evidence that electrical effects could impact the efficiency of coal dust collection by surfactant-

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containing sprays. Page [9] found that coal dust particles could carry varying electric charges after pulverization, and that the amount of charge depended on the characteristics of individual coal mines. The factors affecting the sign and magnitude of charges on coal dust particles include moisture content, ambient humidity, dust components (e.g., mineral level, sulfate level), process method, and particle size [9,10]. Coal dust particles with a diameter of $0.5\ \mu\text{m}$ can carry on the order of 10^2 charges per particle [11], and in certain conditions coal dust particles with a diameter of $550\ \mu\text{m}$ can carry on the order of 10^6 charges per particle [12]. However, typical charge levels on respirable coal dust particles are on the order of 10–100 elemental charges per particle, depending on particle diameter [10]. Electrostatic forces are most important among respirable particles, nominally those $< 4\ \mu\text{m}$ in aerodynamic diameter, because the effects of interception and inertial impaction are negligible for many of these particles [13]. Although the primary particle size mode of coal dust by mass in the United States (US) underground coal mines is $\sim 17\text{--}20\ \mu\text{m}$ [14], it is reasonable to investigate the electrical effects on highly-charged coal dust particles within the respirable size range because those particles tend to deposit more and deeper in the lung and cause more severe health effects than larger particles.

To complement the charged coal particles, surfactant-containing spray drops can also carry a large number of electric charges [15]. The sign and magnitude of charge on spray drops vary according to the classification and concentration of surfactant applied. According to Polat et al [15,16], adding anionic surfactant into distilled water increases the fraction of negatively-charged drops. Therefore, the anionic surfactant-containing spray drops tend to have net negative charge. Conversely, cationic surfactant spray drops tend to carry a net positive charge. Nonionic surfactant spray drops tend to carry a small net positive charge.

Based on the fact that both dust particles and surfactant-containing spray drops can carry considerable amounts of electric charge, Tessum et al [17] measured the efficiency with which surfactant-containing sprays collected charged, laboratory-generated, monodisperse polystyrene latex (PSL) particles with diameters of $0.6\ \mu\text{m}$, $1.0\ \mu\text{m}$, or $2.1\ \mu\text{m}$. They found that, in general, nonionic surfactant-containing sprays are most effective for weakly-charged respirable particles, whereas ionic surfactant-containing sprays are most effective for highly-charged respirable particles with opposite charge. The implications of these results on the choice of surfactant in charged respirable dust suppression are important. However, owing to the difference in size distribution, physical properties, and electrical properties, the collection efficiency of charged, polydisperse coal dust particles by surfactant

spray may differ from the collection efficiency for PSL particles. The objective of this study was to investigate whether adding surfactants into spray water could increase the dust capture efficiency of respirable coal dust particles.

2. Materials and methods

The experimental set-up included particle generation, spray, and particle measurement sections in a wind tunnel, as described in Tessum et al [17]. The wind tunnel has a square cross section of $0.3\ \text{m} \times 0.3\ \text{m}$. The air velocity in the wind tunnel was $0.61\ \text{m/s}$. The apparatus used in this study, shown in Fig. 1, differed from that of Tessum et al [17] in that in order to generate polydisperse coal dust particles of a wide size range, coal dust collected from ground bituminous coal (Austin Black 325, Coal Fillers Inc. Bluefield, VA, USA) was aerosolized in the particle generation section using a Model 3,400 Fluidized Bed Aerosol Generator (TSI Inc., Shoreview, MN, USA) at an air pressure of 40 psi and a 20% bead purge flow rate. The aerosol then either remained unneutralized or was given a neutral, net positive, or net negative charge. An Aerosol Neutralizer (Model 3,012A, TSI Inc.) with a Kr-85 beta particle radiation source was used to imbue the aerosol with a Boltzmann equilibrium charge distribution, i.e., a neutral charge. Then, the neutralized aerosol was optionally mixed with either positive or negative ions generated by high voltage applied to a corona needle in a diffusion charger (taken from a TSI Inc. AeroTrak Nanoparticle Aerosol Monitor 9,000) connected to a regulated DC power supply (Eisco Labs, Ambala Cantt, India). Neutralized aerosol passed through the diffusion charger operating with a voltage of either $-5.3\ \text{kV}$ or $+5.3\ \text{kV}$ and at a flow rate of approximately $4\ \text{L/min}$.

During particle generation, the Fluidized Bed Aerosol Generator was run with a 55% bed flow rate and a 17% bead speed to generate unneutralized and neutralized aerosols, and was run with a 9% bed flow rate and a 57% bead speed to generate positively- and negatively-charged aerosols.

In the spray section, particles were removed as they passed through sprays with plain water or anionic (sodium dodecyl sulfate, SDS, Fisher Scientific Inc., NJ, USA), cationic (dodecylamine hydrochloride, DAH, Tokyo Chemical Industry Co., Japan), or nonionic (Triton X-100, Fisher Scientific Inc., NJ, USA) surfactants in the wind tunnel. SDS releases sodium ions (Na^+) into solution, and thus the anionic spray tends to carry a net negative charge, whereas the DAH spray tends to carry a net positive charge owing to released chloride ions (Cl^-). Different spray solutions with $1 \times 10^{-4}\ \text{M}$ concentration of one of the surfactants were sprayed into the wind tunnel through three lined hollow cone spray nozzles (No. TTD4-46, Spraying

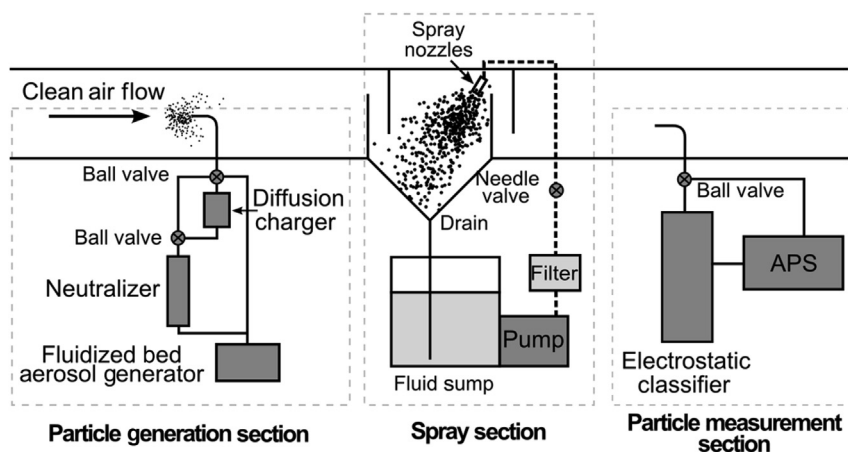


Fig. 1. Schematic diagram of experimental set-up in wind tunnel. APS, aerodynamic particle sizer.

Systems Co., Wheaton, IL, USA). The spray nozzles were operated at a pressure of 552 kPa, a fluid flow rate of 2.95 L/min, and a fixed angle of 33° from vertical. Spray solutions were recirculated and stored in a 189-L sump, and fine particles were filtered out before the solutions were sprayed back into wind tunnel to prevent particle regeneration. The relative humidity in the tunnel at the downstream to spray section was consistently between 95% and 100% during all experiments. This high humidity minimized evaporation of spray drops, and particles did not grow by condensation because the aerosol was not supersaturated with water vapor.

In the particle measurement section, some of the remaining particles were sampled through a subsokinetic probe, optionally selected by a modified aerosol electrostatic classifier (Model 3,071A, TSI Inc.) according to their electrical mobility, and then counted and sized by an aerodynamic particle sizer (APS, Model 3,321, TSI Inc.). In order to measure the sizes and charge levels of coal dust particles in this experiment, an external high voltage DC power supply with a range of 0–±20kV (Bertan 230 High Voltage Power Supply, Spellman High Voltage Electronics Corporation, Hauppauge, NY, USA) replaced the internal voltage supply on the modified aerosol electrostatic classifier to allow mobility-based separation of either positively- or negatively-charged particles. In addition, the flow rate through the classifier was modified to a sheath air flow rate of 10 L/min and an aerosol flow rate of 2 L/min to provide the particle size and charge ranges necessary for this study. Although the user manual for the electrostatic classifier indicates a maximum measurable particle size of 1.0 μm, this maximum size is based on the limit of having single charges on the particles being measured and the standard flow rate through the classifier. Our modifications to the classifier coupled with the size measurements using the APS allowed us to measure the size and charge of particles as large as 5 μm across the full voltage range of the modified classifier. Coal dust concentrations measured by the APS were < 2,700 particles/cm³ for all experimental conditions and at all size bins, which is within the recommended APS concentration limit of 10,000 particles/cm³. As the lower limit of detection of the APS is 0.5 μm, any particles with a smaller diameter are not reported here.

The number of elementary charges on the particle, n , as an indicator of charge level, was calculated as described by Tessum et al [17]:

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

where Z is particle electrical mobility, μ is gas viscosity, D_p is particle diameter, e is elementary charge, and C is Cunningham slip correction. Particle electrical mobility Z also depends on the voltage applied to the electrostatic classifier. Collection efficiencies for particles with all charge levels together were measured when sampled particles were directly counted by the APS without being selected by the electrostatic classifier. Collection efficiencies for particles with specific charge levels were measured when sampled particles were selected by the electrostatic classifier and then counted by the APS. The APS was operated for a 60-second particle count time with an aerosol flow rate of approximately 1 L/min for each measurement.

Collection efficiencies based on net particle count when spray was on/off were calculated from the coal dust penetration, as described by Tessum et al [17], as

$$\eta = 1 - \frac{C_{1,1} - C_{0,1}}{C_{1,0} - C_{0,0}} \quad (2)$$

where η is collection efficiency, C is total particle/drop count by the APS, the first subscript is for particle generation on/off and the

second subscript is for spray on/off, and 1 equals to “on”, and 0 equals to “off”. For each test, the APS was used to measure the four concentrations in Eq. (2). The sequence for the concentration measurements was $C_{0,0}$, $C_{1,0}$, $C_{1,1}$, and then $C_{0,1}$. Each concentration was measured with five repetitions. After each change in the state of particle generation and spray, a 2–3-minute wait was required for the system to reach a steady state before being measured by the APS. To make sure that coal dust particles were generated consistently, the air velocity in the wind tunnel was measured and found to be consistent when spray was or was not applied. As the aim of this study was to compare the performance of different surfactants against each other rather than to measure absolute collection efficiency for our experimental configuration (which could vary with choices of spray nozzle type, nozzle angle, liquid flow rate, air flow rate, etc.), and because the different surfactant solutions will result in similar wall losses, wall losses were not assessed separately in this study. In addition, an analysis of expected wall deposition velocities versus residence time in the apparatus indicates that wall losses were likely negligible for the sizes of coal dust particles evaluated in this study.

As occupational exposure limits for respirable coal dust are based on mass concentration, the respirable mass concentration of coal dust aerosol generated in this study was calculated. Number concentrations measured by the APS in a total of 51 size bins between 0.542 μm and 19.81 μm were used to calculate respirable mass concentration, C_r , as

$$C_r = \sum_{i=1}^{51} C_{n,i} \rho \frac{\pi d_i^3}{6} f_i \quad (3)$$

where $C_{n,i}$ is particle number concentration in size bin i (#/m³), ρ is the density of coal dust, d_i is the geometric mean diameter of each size bin, and f_i is the fraction of respirable aerosol in size bin i (based on aerodynamic diameter) as calculated per the American Conference of Governmental Industrial Hygienists (ACGIH)’s respirable sampling criteria [18]. Coal dust used in this study had a density of 1,310 kg/m³. A dynamic shape factor of 1.05 was used to determine the geometric diameter from aerodynamic diameter for each size interval [19]. The respirable mass collection efficiencies were determined in a similar manner to number efficiencies but using size-integrated respirable mass concentration rather than size-specific number concentration.

In this study, three types of collection efficiency are calculated. Collection efficiencies are aggregated across all charges and sizes are referred to as “overall collection efficiency”. Collection efficiencies that are disaggregated by particle size but aggregated across all particle charges are referred to as “size-specific collection efficiency”. Finally, collection efficiencies that are disaggregated by particle charge but aggregated across all particle sizes are referred to as “charge-specific collection efficiency”.

Controlled conditions for each independent variable in three types of collection efficiency measurements are shown in Tables 1 and 2. The factors tested in this study that may impact the overall and size-specific collection efficiency are aerosol charge condition and surfactant, and factors that impact the collection efficiency for particles with a specific charge level are particle charge level and surfactant. The particle charge levels shown in Table 2 were based on the aerosol charge condition: unneutralized, neutralized, negative, or positive. As shown in Table 2, four positive and four negative voltages were applied to the electrostatic classifier for each charge condition to allow efficiency measurements for eight particle charge levels at each particle size. Each set of measurements was repeated three times. Voltage level, rather than number of charges per particle, is shown in Table 2 because the number of charges per particle varies with particle size.

Table 1
Test conditions for overall and size-specific collection efficiency measurement

Independent factors	Test condition
Aerosol charge condition	Unneutralized Neutralized Negative Positive
Surfactant	10 ⁻⁴ M Anionic (SDS) solution 10 ⁻⁴ M Nonionic (Triton X-100) solution 10 ⁻⁴ M Cationic (DAH) solution Deionized water

DAH, dodecylamine hydrochloride; SDS, sodium dodecyl sulfate.

Table 2
Test conditions for charge-specific collection efficiency measurement

Independent factors	Test condition
Surfactant	10 ⁻⁴ M Anionic (SDS) solution 10 ⁻⁴ M Nonionic (Triton X-100) solution 10 ⁻⁴ M Cationic (DAH) solution Deionized water
Particle charge level	Unneutralized Neutralized Negative/positive ± 8,100 V ± 8,100 V ± 2,430 V ± 2,970 V ± 4,466 V ± 1,331 V ± 1,089 V ± 2,430 V ± 729 V ± 399 V ± 1,331 V ± 399 V

DAH, dodecylamine hydrochloride; SDS, sodium dodecyl sulfate.

The effects of aerosol charge condition and surfactant on overall respirable mass collection efficiency and size-specific number collection efficiency were estimated statistically using a two-way analysis of variance (ANOVA). Scheffé's method [20] was used to evaluate the effect of surfactant on the arithmetic means of size-specific collection efficiency at each aerosol charge condition. The effects of particle charge level and surfactant on charge-specific collection efficiency were estimated using a two-way analysis of variance. Regression analysis was used to predict the relationship between particle charge and collection efficiency for each aerosol charge condition. The arithmetic mean and standard deviation of fractions of total generated coal dust particles with specific charges were analyzed for particles with diameters of 0.6 μm and 1.0 μm in order to compare with previous PSL particle measurements. Statistical analyses were performed using R version 2.13.0 [21].

3. Results

3.1. Coal dust particle size and charge description

As measured by the APS, the geometric mean of the coal dust particle size distribution by number was 0.89 μm with a geometric

standard deviation of 1.42. The geometric mean of the coal dust particle size distribution by mass was 1.51 μm with a geometric standard deviation of 1.64. The coal dust size distributions by number and mass are shown in Fig. 2.

Coal dust particle charge distribution is shown in Fig. 3, Panels A1–A4. Both unneutralized and neutralized coal dust aerosols contained positively and negatively charged particles. Unneutralized aerosol had a net positive charge, whereas neutralized aerosol had a net negative charge. Compared with unneutralized aerosol, neutralized aerosol had a smaller proportion of charged particles and had fewer charges per particle, with a charge distribution close to a Boltzmann charge distribution. Positively- and negatively-charged aerosols had similar numbers of charges per particle with only unipolar charges. However, the positively-charged aerosol had a higher proportion of charged particles than the negatively-charged aerosol, indicating that the positively-charged aerosol had higher net charge.

Fig. 4 shows the fraction of particles carrying electric charges at eight tested charge levels for coal dust particles with 0.6 μm and 1.0 μm diameters (the black dash lines), normalized by the width of the charge interval represented by the measurement. In general, coal dust aerosols had a larger fraction of charged particles for 1 μm particles than for 0.6 μm particles. In addition, unneutralized coal dust aerosol had a small net positive charge and neutralized coal dust aerosol had a small net negative charge. Positively-charged coal dust aerosols had higher net charges than the comparable negatively-charged aerosols.

3.2. Size-specific and overall collection efficiency

Particle size is the most important factor affecting spray efficiency ($p < 0.0001$), therefore, it is most useful to compare collection efficiency at specific particle sizes. Size-specific collection efficiency for coal dust based on particle number concentration increased as particle size increased: 28.8% \pm 18.3%, 65.7% \pm 9.1%, and 87.8% \pm 5.7% (mean \pm SD) for particles 0.6 μm , 1.0 μm , and 2.1 μm in aerodynamic diameter, respectively, averaged across aerosol charge conditions and surfactants. The size-specific number efficiency for coal dust particles for each charge condition is shown in Fig. 5. Larger variances of size-specific collection efficiency are observed among negatively- and positively-charged aerosols. The negative and positive aerosols lost many particles during diffusion charging, leading to fewer particles and greater variance during efficiency measurements.

Across all four sprays, the average respirable mass collection efficiency for coal dust was 75.5 \pm 9.4% [mean and 95% confidence interval (CI)]. The average overall respirable mass collection efficiency for coal dust was 69.0 \pm 7.6%, 76.2 \pm 6.9%, 78.0 \pm 7.6%, and

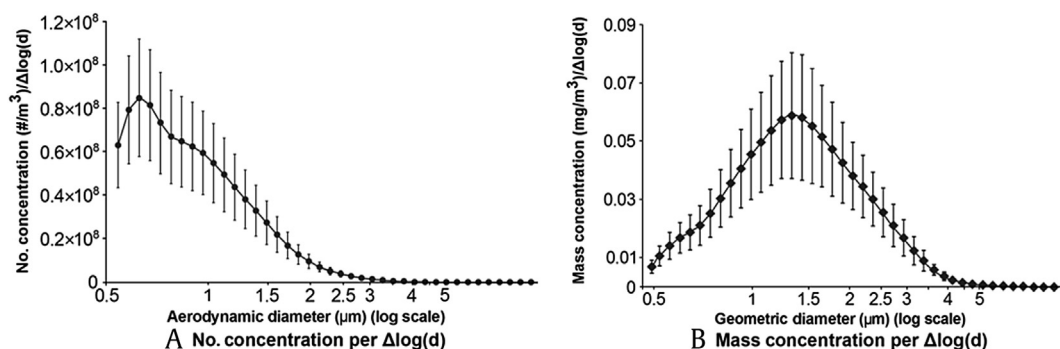


Fig. 2. Coal dust aerosol particle concentration per $\Delta\log(d)$ (mean \pm SD, $n = 12$) as a function of particle size for (A) number concentration and (A) mass concentration. SD, standard deviation.

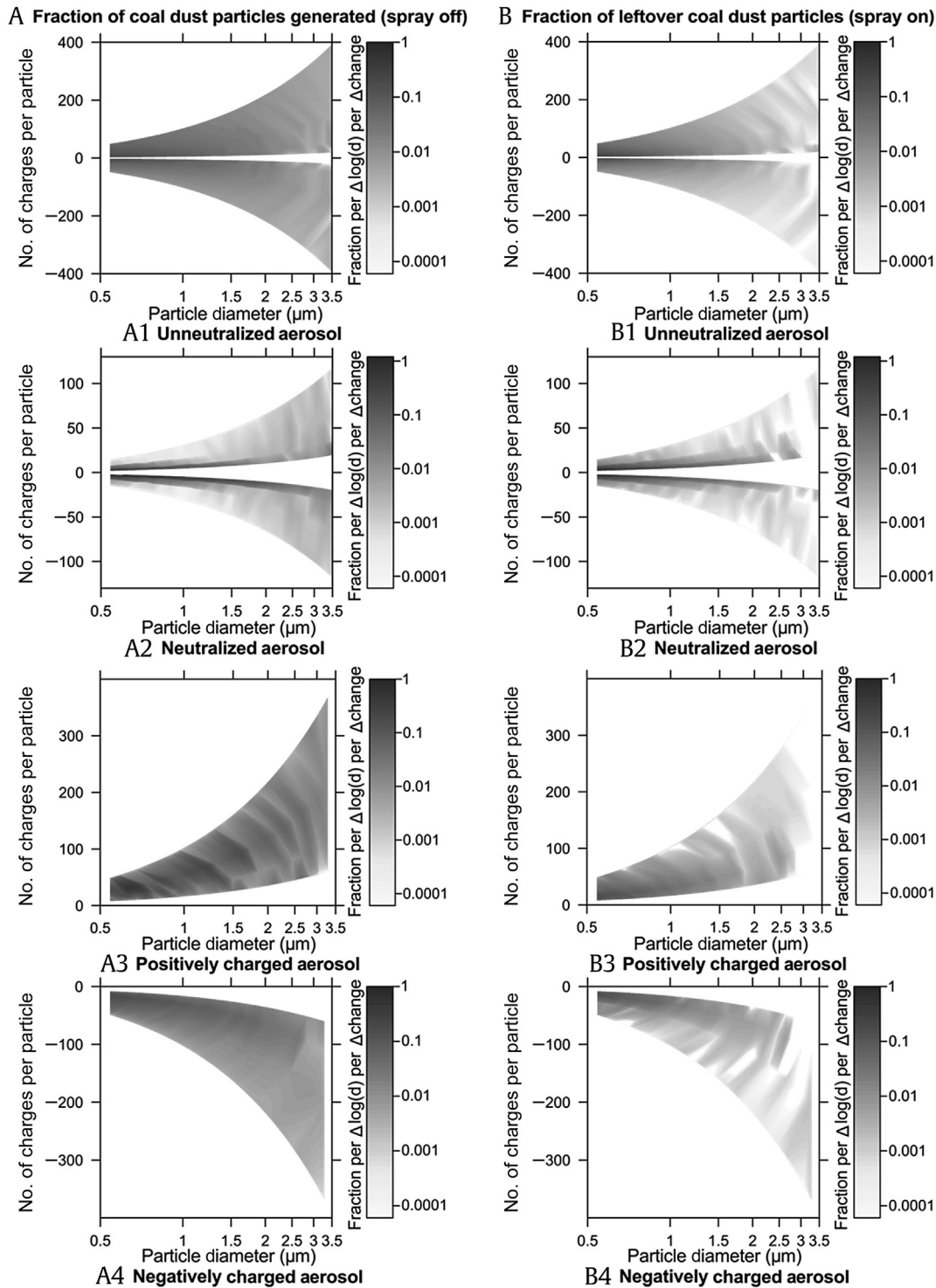


Fig. 3. Mean fractions of charged coal dust particles at different particle sizes and charge levels when (A) spray is off and (B) when spray is on for four (1–4) aerosol charge conditions, normalized by both $\Delta\log(d)$ and $\Delta(\text{number of charges})$. Values in panels B1–4 represent averages among all three surfactants and plain water.

$78.8 \pm 7.1\%$ (mean and 95% CI) for the anionic, cationic, nonionic surfactant spray, and water spray, respectively. A two-way ANOVA test suggested that the overall respirable mass efficiency was significantly impacted by the use of surfactant ($p < 0.0001$), and that anionic surfactant spray had a significantly lower average respirable mass collection efficiency than other surfactants. The aerosol charge condition and the interaction of surfactant and aerosol charge condition did not affect overall collection efficiency ($p = 0.875$ and $p = 0.112$, respectively).

Although the interaction of surfactant and aerosol charge condition did not significantly impact overall respirable mass collection efficiency, the effect of surfactant was different among different aerosol charge conditions for the count-based collection efficiency that varied with particle diameter. For example, surfactant did not significantly impact number collection efficiency for unneutralized, and positively-charged aerosols, but significantly impacted collection efficiency for neutralized and negatively-charged aerosols according to Scheffé's test, as shown in Fig. 6. Overall, the nonionic

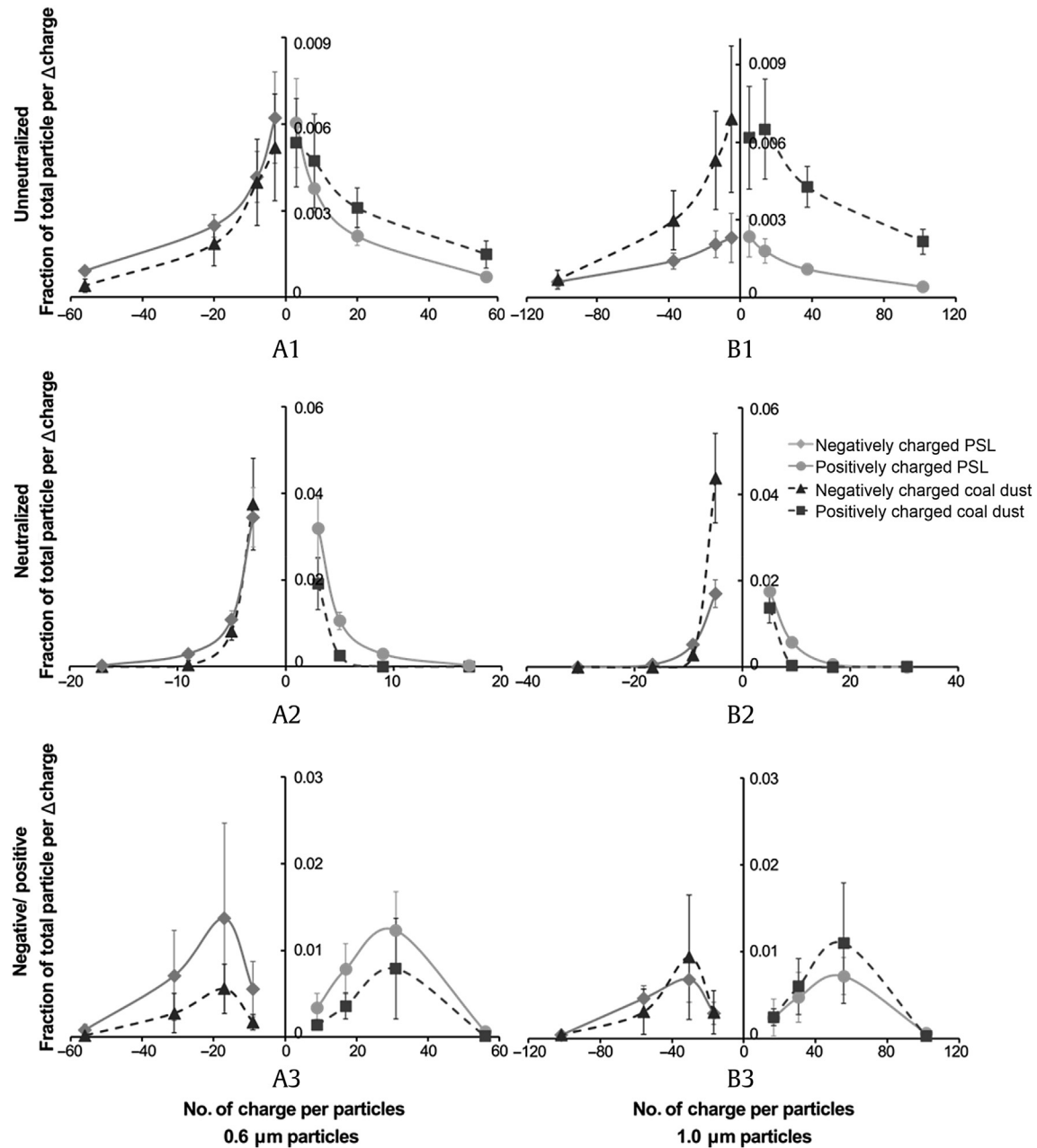


Fig. 4. Comparison of fractions of total generated particles (mean \pm SD) of PSL and coal dust at different charge levels for (A) 0.6 μm and (B) 1.0 μm particle size for four aerosol charge conditions: Unneutralized particles are shown on the top row (A1 and B1), neutralized particles are shown in the middle row (A2 and B2), negatively and positively charged particles are combined in the bottom row (A3 and B3). The fraction was calculated by the number of 0.6 μm or 1.0 μm particles with a given charge divided by the total number of generated particles of the same size, and normalized by Δ (number of charges). PSL, polystyrene latex; SD, standard deviation.

surfactant tended to have the same or higher collection efficiency on neutralized and negatively-charged aerosols relative to the other surfactants, whereas anionic surfactant tended to have the same or lower efficiency than the other sprays. Larger variances of size-specific collection efficiency are observed among negative and positive aerosol charge condition, which may mask some of the impact on size-specific collection efficiency by surfactants.

3.3. Collection efficiency for particles with specific charges

As particle size influences the number of charges a particle can carry, as shown in Fig. 3, charge-specific collection efficiencies were analyzed at specific size levels. Both surfactant and particle charge level can impact spray collection efficiency. A two-way ANOVA of

charge-specific collection efficiencies suggests that particle charge level was a more important factor than surfactant in impacting charge-specific collection efficiency at specific particle sizes. However, the interaction of particle charge level and surfactant also significantly affected charge-specific collection efficiency. This result indicates that particle charge level influences charge-specific collection efficiency regardless of the type of surfactant used, but surfactant substantially influences the charge-specific collection efficiencies for particles at certain charge levels. For example, surfactant did not significantly affect the collection efficiency for 1 μm particles of positively-charged aerosol ($p = 0.917$), but both the particle charge level and the interaction of particle charge level and surfactant did have significant impact ($p = 0.0002$ and $p = 0.0035$, respectively), as shown in Table 3.

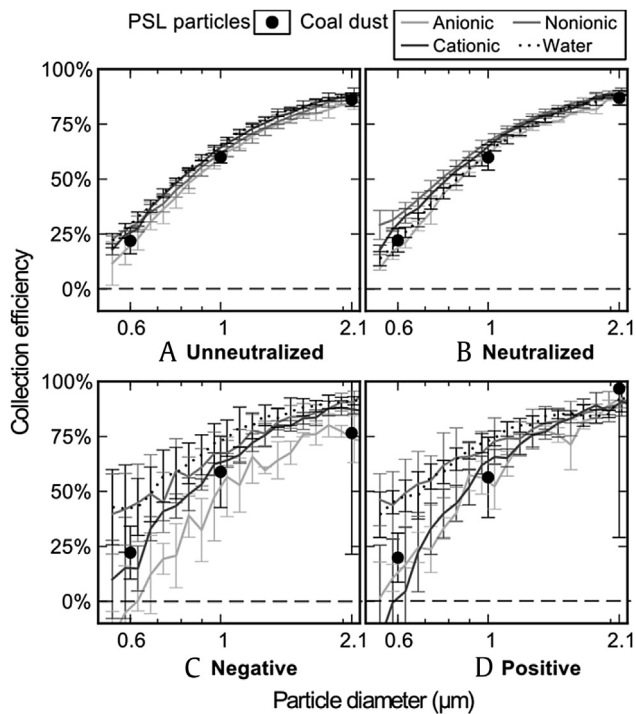


Fig. 5. Size-specific overall collection efficiency (mean \pm SD) of coal dust and PSL particles as a function of particle diameter for four aerosol charge conditions. The overall collection efficiencies of PSL particles were averaged across different types of spray. PSL, polystyrene latex; SD, standard deviation.

Comparing the leftover coal dust particles (Fig. 3B) to total generated particles (Fig. 3A) shows that particles with larger size were removed more efficiently than smaller particles, and highly-charged particles were removed more efficiently than weakly-charged particles across all surfactant-containing sprays and plain water spray. These observations are also shown among unneutralized aerosols in the three particle sizes in Fig. 7. Higher efficiencies were also seen among highly-charged particles in positively- and negatively-charged aerosols. However, this trend cannot be observed for neutralized aerosols owing to the limited number of charged particles that were generated.

4. Discussion

The two major factors that were observed in this research to substantially drive the electrical effects on dust capture were particle size and particle charge. Smaller and more highly charged particles were influenced more by charged spray drops, whereas coal dust particles with particle diameter $> 2.1 \mu\text{m}$ were affected much less by charge effects (Fig. 7). It is worth noting that particles with high charge also cause more severe health effects: Melandri et al [22] demonstrated that charged particles exhibited greater deposition in the lungs than particles that were neutralized. Therefore, use of electrical effects to control charged particles could lead to greater health benefits than would techniques designed to control all particles.

The spray collection efficiencies on monodisperse PSL particles with different charge conditions were discussed in our previous article [17]. Comparing the charge distribution of coal dust and PSL particles at each charge condition (Fig. 4), we found that PSL aerosol had a larger fraction of charged particles at each charge level for $0.6 \mu\text{m}$ particles than for $1 \mu\text{m}$ particles, whereas coal dust aerosols had a larger fraction of charged particles for $1 \mu\text{m}$ particles than for

$0.6 \mu\text{m}$ particles. In general, compared with PSL aerosols at each aerosol charge condition, coal dust aerosol had a similar or lower proportion of charged particles and lower charge levels for $0.6 \mu\text{m}$ particles, especially for positively- and negatively-charged aerosols, but had a similar or higher proportion of charged particles and lower charge levels for $1.0 \mu\text{m}$ particles. These differences were statistically significant, indicating that coal dust tends to have weaker charge properties in both quantity and magnitude for smaller particles. As more than half of measured coal dust particles were $< 0.8 \mu\text{m}$ (Fig. 2A), the charge properties of coal dust aerosols across all particle sizes are weaker on average than those of PSL aerosols, especially for positively- and negatively-charged aerosols.

Our previous research has found that ionic surfactant-containing spray enhances dust capture for particles with a highly opposite charge [17]. However, ionic surfactant-containing sprays did not show significant enhancement in charged coal dust particle capture in this study. The only significant impact of surfactant use is that anionic surfactant-containing spray had similar or lower efficiency on negatively-charged coal dust particles compared with other surfactants. This may be because, compared with the PSL particles, the coal dust particles generated in this study had a lower fraction of highly-charged particles, especially for both positively- and negatively-charged aerosols, indicating that the electrical effects may only play a significant role in highly-charged particle control. Therefore, the impact of ionic surfactants may be more noticeable for aerosols with high charge level.

Although information regarding the difference between electrical properties of PSL spheres and coal dust is limited, one study has summarized that when both polystyrene and coal dust aerosol were generated with Wright-type nebulizer and dust feeder, the coal dust carried more charges than did the polystyrene particles of the same particle size [10]. In our study, there are several possible causes for fewer particle charges observed in positively- and negatively-charged coal dust aerosol as compared with PSL: (1) compared with the PSL particles, the coal dust particles went through the diffusion charger at a higher concentration, which may have caused each particle to gain fewer ions; and (2) the flow rate of coal dust aerosol entering the diffusion charger was $\sim 4 \text{ L/min}$, which was higher than the 2.3 L/min flow rate of the PSL aerosol. This difference caused the coal dust particles to have less residence time, on average, to obtain charges within the diffusion charger. Although the coal dust particles had limited charge numbers in this study, coal dust particles may carry more charges and be more sensitive to charged spray drops in actual coal mines. Information comparing electrical charge on coal dust aerosol generated in actual mining conditions and in laboratories is limited, but Johnston et al [10] reported the charge measurements in a coal mine by a Russian study. The charge distribution observed in the Russian coal mine showed that proportions of negatively- and positively-charged particles were similar, and most particles had small amounts of charge. However, the maximum charge was > 300 charges per particle in the Russian study, which is much higher than our laboratory-generated coal dust particles. As there was no particle size information given by Johnston et al [10], it is not possible to make a robust charge comparison. Nonetheless, coal dust particles in underground coal mines are larger in diameter and are more likely highly charged on average than the particles generated for this study.

Undoubtedly, surfactant use affects spray efficiency. Although surfactants did not significantly impact size-specific collection efficiency for all aerosol charge conditions, nonionic surfactant sprays had better overall performance on weakly charged neutralized aerosols and on negatively-charged aerosols, which had fewer charges per particle compared with the unneutralized and positively-charged aerosols (Fig. 3). The observation that nonionic

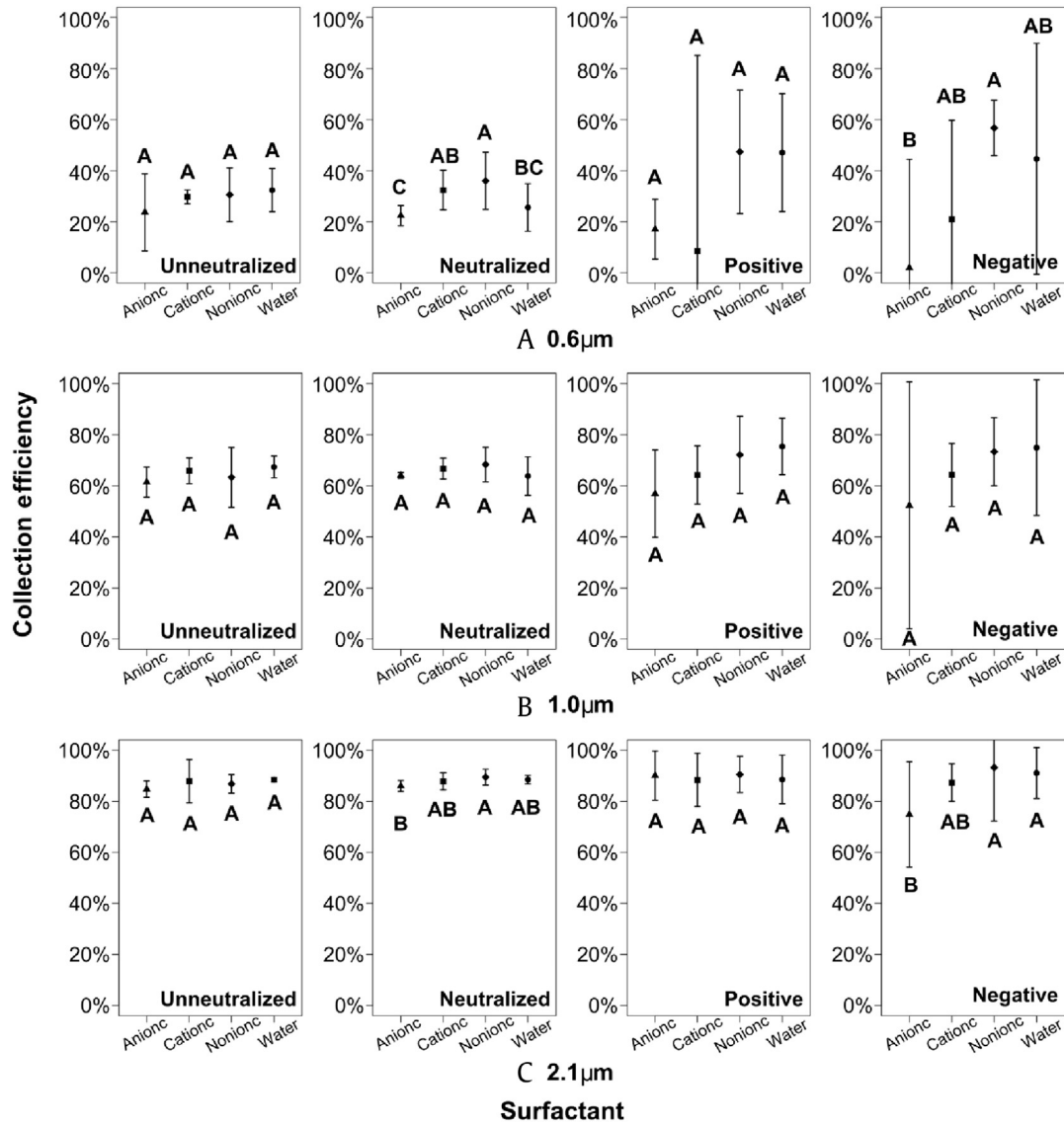


Fig. 6. Mean size-specific overall number collection efficiency with 95% confidence intervals as a function of surfactant for four aerosol charge conditions for three particle sizes at 0.6 μm, 1.0 μm and 2.1 μm. Within each figure, means with different letters are significantly different (Scheffé test, $p < 0.05$). PSL, polystyrene latex; SD, standard deviation.

Table 3
Significance results for two-way ANOVA for the impact of surfactant and particle charge level on collection efficiency

Test condition		Two-way ANOVA result (p)		
Particle size	Aerosol charge condition	Surfactant	Particle charge level	Interaction
0.6 μm	Positive	0.475	0.810	0.138
	Negative	0.938	0.197	0.266
	Neutralized	< 0.0001*	< 0.0001*	< 0.0001*
	Unneutralized	0.874	0.314	0.022*
1.0 μm	Positive	0.917	0.0002*	0.004*
	Negative	0.482	0.056	0.327
	Neutralized	0.017*	0.0001*	< 0.0001*
	Unneutralized	0.466	0.088	0.290
2.1 μm	Positive	0.141	0.065	0.338
	Negative	0.329	0.865	0.040*
	Neutralized	0.004*	< 0.0001*	< 0.0001*
	Unneutralized	0.973	0.563	0.786

* $p < 0.05$.
ANOVA, analysis of variance.

surfactant can significantly remove weakly-charged particles concurs with previous PSL results [17], indicating that, besides electrical effects, it is possible that there may be other mechanisms influencing dust capture such as surface tension and drop diameter. According to Tien and Kim [4], drops with lower surface tension tend to have less resistance to the sorption of hydrophobic coal particles, and thus have a better capacity to capture airborne particles. Lower surface tension may also decrease the diameter of spray drops, and this change in drop size distribution may lead to a higher spray efficiency [23]. Future analyses of the effects of surfactants on collection efficiency should consider the impacts of surface tension and spray drop size.

One limitation of this study is that we have assumed that the sign and magnitude of charges on spray drops in our experiments are the same as those reported by Polat et al [16]. Polat et al [16] found that, for a given surfactant concentration, ionic surfactant-containing sprays tend to carry a net corresponding charge and nonionic surfactant (Triton X-100) spray tends to have weak positive charge. They also found that plain water has an even weaker positive charge than nonionic surfactant and that adding ionic

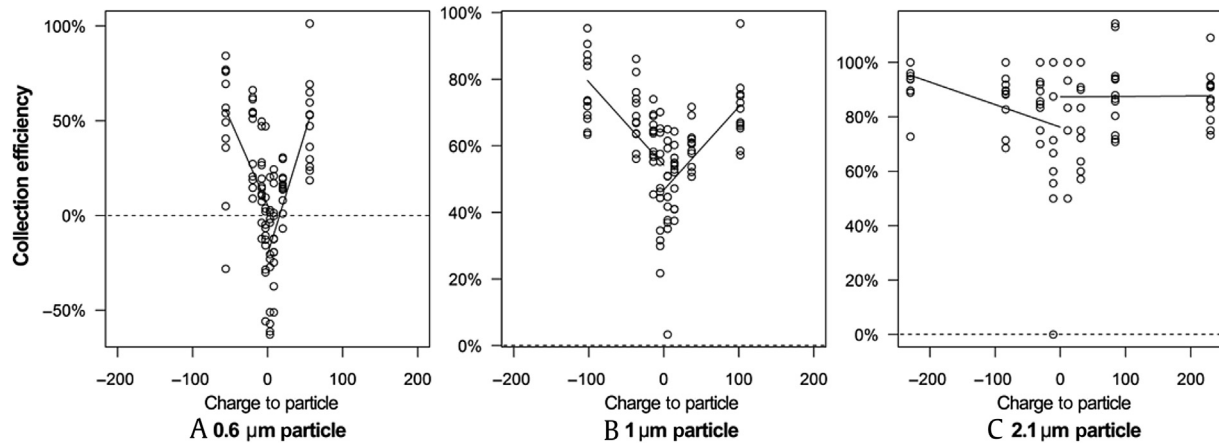


Fig. 7. Scatter plot with regression line of collection efficiency on unneutralized coal dust aerosol as a function of particle charge level for three particle sizes at 0.6 μm , 1.0 μm and 2.1 μm .

surfactants substantially enhances the charge level of spray drops. However, differences in our experimental setup relative to that of Polat et al [16] (e.g., nozzle type, flow rate of the spray solution, and pressure applied on nozzle) may have caused the charge properties of the spray drops in our experiments to be different than those reported by Polat et al [16]. Further investigation is required to determine whether differences in experimental conditions affect drop charge characteristics.

Another limitation in this study is that the measurements of charge-specific collection efficiency have some inaccuracies. For example, negative collection efficiencies were observed among weakly-charged 0.6 μm particles (Fig. 7). As discussed in Tessum et al [17], when the APS was used to count particles at each size and certain charge levels, the APS could not provide information regarding whether the counted particles were coal dust, drops, or a combination of the two. Spray drops at respirable sizes tended to be almost neutrally charged. Coal dust, especially in the unneutralized and neutralized aerosols, had many particles with charge levels lower than we could measure, as shown in Fig. 4. However, if any spray drops, coal dust, or combined particles with these lower charge levels gained charge as they traveled through the particle measurement section, they would be measured by the APS and be counted as our targeted charged coal dust, causing an increase in $C_{1,1}$, in Eq. (2), and resulting in the underestimation of charge-specific collection efficiency.

Theoretically, the charging of spray drops and coal dust with lower charge levels in the experimental apparatus could occur in two ways: (1) drop evaporation may cause charging as described by Lear and Harmon [24]. When large, highly-charged drops partially evaporate while traveling with the air flow in a wind tunnel and their size decreases (but charge stays the same), drop charge level can approach the Rayleigh limit and charges can be released by one of several instability mechanisms. The charge released by an evaporated drop can be transferred to nearby drops or coal dust. In our case, drops and coal dust would be more likely to gain charges when they were almost neutral or only had fewer charges, and this may be the reason that the collection efficiencies of weakly-charged particles were more susceptible to this phenomenon; and (2) the charging of spray drops and coal dust with lower charge levels may have happened when charged coal dust collided with spray drops, which may have caused the charges on the drop and coal dust to combine. For example, a charged coal dust particle with diameter $< 0.6 \mu\text{m}$ could collide with a neutral drop also having a diameter $< 0.6 \mu\text{m}$, and the combined particle and drop could be

measured by the APS as a charged 0.6 μm particle. Although these mechanisms could cause the underestimate of charge-specific collection efficiency, this underestimate would be consistent among all tested sprays, and therefore likely had limited impact on our analyses of advantages of a particular surfactant.

5. Conclusion

Respirable coal dust particle size is positively associated with spray collection efficiency, significantly higher efficiencies are observed among larger particles in all test conditions. All surfactant-containing sprays and plain water spray tested in this study showed similar overall performance in capturing highly charged unneutralized aerosol and positively-charged aerosol. However, the nonionic surfactant-containing spray had the same or significantly better size-specific collection efficiency and charge-specific collection efficiency for weakly-charged neutralized and negatively-charged coal dust aerosols relative to other sprays. This observation may have limited importance in actual mining conditions because coal dusts generated in coal mines are likely to carry much higher charge level than the ones generated in our lab. Charge-specific collection efficiency tests suggest that smaller respirable coal dust particles ($< 2 \mu\text{m}$) were more sensitive to the surfactant used in a spray, especially among highly-charged particles, indicating a potential that ionic surfactants may have an enhanced performance in highly-charged particles of the opposite sign. Spray efficiency results demonstrate that the overall capture of coal dust particles by spray is similar to the capture of equivalent-sized monodisperse PSL particles, suggesting that PSL particles can be a reasonable surrogate for coal dust in future overall and size-specific collection efficiency studies. However, because PSL and coal particles have different charge capacities, care must be taken when using PSL particles to represent coal dust when performing collection efficiency tests involving electrical effects.

Conflicts of interest

The authors have no conflicts of interest to declare.

Acknowledgments

The study in this publication was supported by CDC-NIOSH Grant (Number 1R01OH009768). Its contents are solely the responsibility of the authors and do not necessarily represent the official views of

NIOSH or CDC. The authors would like to acknowledge Coal Fillers Inc. for supplying the coal dust used in this study.

References

- [1] National Institute for Occupational Safety and Health (NIOSH). Work-related lung disease surveillance report 2007. Morgantown (WV): National Institute for Occupational Saf and Health (US); 2008. Report No.: Pub. No. 2,008–143a.
- [2] Kobrick T. Water as a control method, state of the art, sprays, and wetting agents. Proceedings of the symposium on respirable coal mine dust. Washington, D.C., US: Bureau of Mines; 1970. Report No.: Information Circular 8,458. p.123–32.
- [3] Chander S, Mohal BR, Aplan FF. Wetting characteristics of particles and their significance in dust abatement. Proceedings of Respirable Dust in the Mineral Industries: Health Effects, Characterization and Control. PA: University Park; 1988. p. 19–23.
- [4] Tien JC, Kim J. Respirable coal dust control using surfactants. *Appl Occup Environ Hyg* 1997;12:957–63.
- [5] Kim J. Respirable coal dust control using surfactants—with special emphasis on liquid spray system [PhD Dissertation]. Rolla (MO): Department of Mining Engineering, University of Missouri-Rolla; 1995.
- [6] Hirschi JC, Chugh YP, Saha A, Mohany M. Evaluating the use of surfactants to enhance dust control efficiency of wet scrubbers for Illinois coal seams. Proceedings of the North American/9th US mine ventilation symposium. Lisse (Netherlands): Balkema; 2002. p. 601–6.
- [7] Kilau HW, Lantto OL, Olson KS, Myren TS, Volt JI. Suppression of longwall respirable dust using conventional water sprays inoculated with surfactants and polymers. US: Department of the Interior, Bureau of Mines; 1996. Report No.: PB96–137328/XAB, RI 9591.
- [8] Polat H, Hu Q, Polat M, Chander S. The effect of drop and particle charge on dust suppression by wetting agents. Proceedings of the 6th US mine ventilation symposium. Littleton (CO): Society for Mining, Metallurgy, and Exploration, Inc.; 1993. p. 535–41.
- [9] Page SJ. Relationships between electrostatic charging characteristics, moisture content, and airborne dust generation for subbituminous and bituminous coals. *Aerosol Sci Tech* 2000;32:249–67.
- [10] Johnston AM, Vincent JH, Jones AD. Measurements of electric charge for workplace aerosols. *Ann Occup Hyg* 1985;29:271–84.
- [11] Walkenhorst W. Charge measurement at dust particles. *Staub-Reinhalt Luft* 1971;31:8–16.
- [12] Nieh S, Nguyen T. Measurement and control of electrostatic charges on pulverized coal in a pneumatic pipeline. *Particul Sci Technol* 1987;5: 115–30.
- [13] Dhariwal V, Hall PG, Ray AK. Measurements of collection efficiency of single, charged droplets suspended in a stream of submicron particles with an electrodynamic balance. *J Aerosol Sci* 1993;24:197–209.
- [14] Burkhart JE, McCawley MA, Wheeler RW. Particle size distributions in underground coal mines. *Amlnd Hyg Assoc J* 1987;48:122–6.
- [15] Polat M, Polat H, Chander S, Hogg R. Characterization of airborne particles and drops: relation to amount of airborne dust and dust collection efficiency. *Part Part Syst Charact* 2002;19:38–46.
- [16] Polat M, Polat H, Chander S. Electrostatic charge on spray droplets of aqueous surfactant solutions. *J Aerosol Sci* 2000;31:551–62.
- [17] Tessum MW, Raynor PC, Keating-Klika L. Factors influencing the airborne capture of respirable charged particles by surfactants in water sprays. *J Occup Environ Hyg* 2014;11:571–82.
- [18] American Conference of Governmental Industrial Hygienists. 2010 TLVs and BEIs based on the documentation of the threshold limit values for chemical substances and physical agents and biological exposure indices. Cincinnati (OH): ACGIH; 2000. p. 1–94.
- [19] Hinds WC. *Aerosol technology: properties, behavior, and measurement of airborne particles*. 2nd ed. New York (NY): Wiley-Interscience; 1999. p. 316–48.
- [20] Scheffé H. A method for judging all contrasts in the analysis of variance. *Biometrika* 1953;40:87–104.
- [21] The R project for statistical computing [Internet]. 2011 [cited 2014 March 3]. Available from: <http://cran.r-project.org/bin/windows/base/old/2.13.0/>.
- [22] Melandri C, Prodi V, Tarroni G, Formignani M, DeZaiacomio T, Bompani GF, Maestri G. On the deposition of unipolar charged particles in the human respiratory tract. In: Walton WH, editor. *Inhaled particle IV*. Oxford (UK): Pergamon Press; 1977. p. 193–201.
- [23] Woffinden GJ, Markowski GR, Ensor DS. Effects of interfacial properties on collection of fine particles by wet scrubbers (EPA-600/7-78-097). Altadena (CA): Meteorology Research Inc.; 1978.
- [24] Lear CW, Harmon DL. EPA Environmental Protection Technology Series: Charged droplet scrubber for fine particle control: Laboratory Study (EPA-600/2-76-249a). [Internet]. Research Triangle Park (NC): US Environmental Protection Agency; 1976 [cited 2015 March 30]. Available from: <http://nepis.epa.gov/Exe/ZyPDF.cgi/910162P6.PDF?Dockey=910162P6.PDF>.