

Effects of Spray Surfactant and Particle Charge on Respirable Dust Control

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Dedication

To Chris, my sea of tranquility.

Abstract

Great efforts have been taken in underground coal mines to prevent coal workers' pneumoconiosis by controlling respirable coal dust, but respirable coal dust levels in mines still often exceed occupational exposure limits. Water-based spray systems are one of the primary dust control methods in mines; studies have suggested a potential to improve spray dust collection efficiency by adding surfactants.

This dissertation investigates the effectiveness of different surfactant-containing sprays in capturing lab-generated monodisperse polystyrene latex particles and polydisperse coal dust by analyzing the impact of particle diameter, aerosol charging condition, surfactant type, sign and magnitude of particle charge, spray solution surface tension, spray drop size, and sign and magnitude of spray drop charge on spray collection efficiency. In order to focus on how the electrical effects caused by adding surfactants to spray water impact spray collection efficiency, the spray surface tension and drop size were taken into account during statistical modeling.

Results indicate that particle size had the most important impact on respirable dust capture by water-based spray. Most particles with a diameter greater than 2 μm can be removed by the spray regardless of other factors. The magnitude of particle charge affected spray efficiency in that highly-charged particles tended to be removed more efficiently than weakly-charged particles.

The magnitude and sign of the charge on surfactant-containing spray drops depended on both surfactant classification and concentration. Pure water spray drops and spray drops with nonionic (Triton X-100) and cationic (DAH) surfactants tended to carry net positive charges on average, whereas spray drops with anionic (SDS) surfactant tended to carry a net negative charge on average. However, the magnitude of

spray drop charge was independent from the concentration of surfactant in the spray water. After controlling the surface tension and drop size, test results indicated a positive correlation between the magnitude of spray drop charge and spray collection efficiency.

Nonionic surfactant-containing spray performed better in capturing respirable dust than the other sprays tested here, especially for weakly-charged aerosols. This superiority may be due to the relatively low surface tension and high charge magnitude of drops containing nonionic surfactant as compared to the other sprays tested here.

Although the predominant mechanisms for respirable dust capture by water-based spray are inertial impaction and interception, electrical effects are also an important factor, especially for highly-charged particles. Therefore, the electrical effects caused by adding surfactants to spray water should be a consideration for future research regarding surfactant effectiveness.

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Chapter 1

Introduction

1.1 Respirable Coal Dust Exposure and Coal Workers' Pneumoconiosis

Mining has been historically considered as the most dangerous occupation in the U.S. and in the world [1]. Coal mining is the biggest mining industry in the U.S., employing 123,259 coal workers in 1,701 coal mines—one third of all miners—in 2013 [2].

Although coal mining accidents, such as explosions, cave-ins, and floods, tend to generate the most news coverage, the greatest occupational hazard in the coal mining industry is chronic respirable coal dust exposure, which causes about six times more total Years of Potential Life Lost (YPLL) before age 65 than fatal injury does, as shown in Table 1.1. Since the Federal Mine Safety and Health Act of 1977 was passed, the Mine Safety and Health Administration has made improvements in decreasing fatalities from Coal Workers' Pneumoconiosis (CWP, or black lung) (Figure 1.1). However, CWP is still a serious occupational health concern. It is a contributing or underlying cause for a total of more than 52,000 coal workers' deaths between 1978 and 2010 [3],

costing about \$39 billion federal compensation [4].

Table 1.1: Fatalities from Injury and from Coal Workers' Pneumoconiosis in U.S. Coal Mines

Health Indicator	Fatal Injury	CWP
Total Fatalities (1978-2010)	2,075[5]	52,149 ^a [3]
Average Annual Fatalities (since 2007)	27[5]	491[3]
Average YPLL-65	28.92 ^{bc} [6]	9.5 ^d [7]
Total Years of Potential Life Lost Before Age 65	780	4660

^aas contributing and underlying causes of death

^bAssume the average YPLL-65 for all types of mining is the same as coal mining

^cUses average YPLL-65 between 1980-1992

^dUses average YPLL-65 between 1968-2006

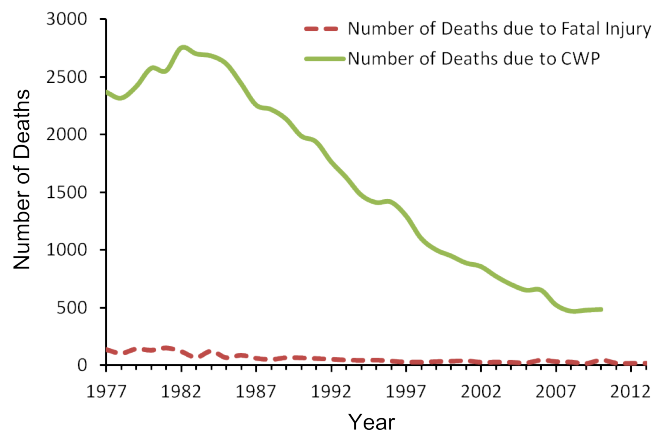


Figure 1.1: Annual fatalities from injury and coal workers' pneumoconiosis, 1977-2013 [5, 3].

In 2014, MSHA lowered the permissible exposure limit (PEL) for respirable coal mine dust from 2 mg/m^3 to 1.5 mg/m^3 , aiming to reduce mortality and morbidity from CWP among coal workers. The equivalent recommended exposure limit (REL) is 1 mg/m^3 , set by National Institute for Occupational Safety and Health (NIOSH) in 1995.

This new federal standard was adopted based on a balance of risk assessment and regulatory economic analysis, and does not mean that respirable coal mine dust exposure level at or below this standard will not cause any adverse health effects. According to MSHA, relative risks of CWP mortality range from approximately 1.6 to 2.1 when the average coal dust exposure is 1.5 mg/m^3 —the current PEL—over a miner's 45-year working history, using the Attfield-Kuempel or P/11 (at age 65, 73, and 80) models [8]. The relative risk of CWP mortality shows a positive correlation with average coal dust concentration, with relative risks ranging from approximately 1.4 to 1.6 when the average dust exposure is at current REL of 1 mg/m^3 , and from approximately 2.0 to 2.7 when the average dust exposure is at the previous PEL of 2 mg/m^3 [8].

Beyond the fact that the occupational exposure limits for respirable coal dust cannot completely eliminate the risk of CWP, the most challenging issue in respirable coal dust exposure is that some coal mines, especially underground coal mines, have difficulty meeting the federal standards. According to MSHA, about 22% of coal mine respirable dust measurements across all occupations among underground coal workers were in exceedance of the 1 mg/m^3 REL, and about 4% of coal mine respirable dust measurements were greater than previous PEL of 2 mg/m^3 based on MSHA inspector samples during 2004 and 2008 [8]. Certain mining operations, such as longwall mining, are associated with higher respirable coal dust concentration than other operations owing to highly mechanized mining methods and higher production levels [10]. For example, about 48% of coal mine respirable dust measurements among longwall miners exceeded the 1 mg/m^3 REL, about 8% of which were in exceedance of the previous PEL of 2 mg/m^3 . Therefore, reducing the respirable coal dust concentrations in underground mines can improve mining health and safety.

1.2 Current Water Spray Dust Control Methods

Water-based spray systems are one of the primary dust control measures in coal mines. Since the 1940s, water sprays have been shown to effectively control dust as compared to dry cutting with an 80% improvement [9]. Water spray systems can effectively reduce the coal dust generation by preventing newly formed dust particles from becoming airborne. Once coal dust is airborne, water spray systems can be used to direct coal dust away from coal workers as a blocking spray [10]. Spray systems can also capture some of the airborne dust by causing the dust to collide with and either adhere to or be completely absorbed by spray drops. The wetted dust then agglomerates and falls from the air [11].

Currently, nearly all longwall mining and continuous mining operations in underground coal mines are equipped with water based spray systems [12, 13]. Although water spray systems are widely used in coal mines, properly designed water spray systems can only reduce respirable dust by about 20 to 60%, with an average of 30% under actual mining conditions [14]. The spray effectiveness at reducing respirable dust is mainly caused by preventing dust from becoming airborne rather than capturing dust in the air. Some studies have referred to dust prevention as primary dust suppression, and airborne dust capture as secondary dust suppression. According to Faschingleitner and Höflinger, a total dust suppression efficiency of 70% was observed on lab generated Pural NF filter test dust with dust size $<10\ \mu\text{m}$ and a mass median of about $4\ \mu\text{m}$, and the efficiency of primary dust suppression (64%) was about 4 times the efficiency of secondary dust suppression (16%) [15]. Because the water spray efficiency for airborne respirable dust capture is not high, there is potential for improvement.

Over the past several decades, scientists have identified factors that impact spray

effectiveness at reducing respirable dust. Well-investigated factors include spray nozzle type, location/position, pattern, flow, and pressure [10]. For the purpose of airborne dust capture, which is the focus of this dissertation, some factors, such as spray nozzle type and pressure, are most important because well-selected nozzle type and pressure can increase airborne respirable coal dust capture by generating high velocity and small-size spray drops [10].

Spray nozzle type can impact airborne dust capture. In general, higher capture efficiencies are seen among nozzles generating smaller drops. Therefore, the dust capture performance of commonly used nozzles, in order from most to least desirable, are 1) air-atomizing nozzles, 2) hollow-cone nozzles, 3) flat-spray nozzles, and 4) full-cone nozzles [10]. However, some studies have also suggested that very small drops can cause captured dust to redisperse owing to evaporation [15, 16]. According to NIOSH, when the sizes of spray drops are similar to dust sizes, in the range of 10 to 150 μm , the spray has the highest efficiency [16].

Spray nozzle operating pressure can affect both drop size and velocity. Higher pressure is associated with smaller drop size and high spray velocity, and therefore increases dust capture effectiveness. Gemci *et al.* have found that doubling the nozzle pressure results in higher velocity, increasing the water flow rate by 23% and decreasing the Sauter mean diameter of spray drops [17]. However, studies have also found that spray pressure above 100 psi can entrain air flow and cause more dust release [10]. In U.S. coal mines, the recommended spray pressure range is 80–100 psi for wetting and dust capturing purposes, and no less than 150 psi for air directing purposes [10].

Other spray characteristics that impact the dust capture, such as nozzle angle, liquid flow rate, distance and location/position, are briefly described in Table 1.2.

Table 1.2: Spray Characteristics and Their Impact on Capturing Respirable Coal Dust

Spray Characterization	Definition	Impact Description
Nozzle Angle	The angle that describes the width of the spray jet as it leaves the nozzle.	High performance associates with wide spray angle [18].
Water Flow Rate	The volume of liquid that passes through the spray nozzle per unit time.	High performance associates with increased liquid water flow [16].
Distance	The distance between major concentrated dust cloud and spray orifice.	Most effective capture within first 12 inch distance[10].
Position	The nozzle placement relative to the dust cloud.	Allows the spray patterns to properly target the concentrated dust flow and at an effective distance that drops will not be carried away by air flow [16].

1.3 Effects of Surfactant Use in Water Spray

In addition to optimizing spray characterization, adding surfactants to spray water is believed to potentially prevent dust generation by reducing the surface tension and increasing the wettability of coal surfaces, thereby improving airborne dust capture [10]. Literature regarding the mechanism by which surfactants improve airborne dust capture will be discussed.

1.3.1 Mechanisms by which Surfactants Affect Dust Capture

Altering Spray Drop Pattern

Since surfactants can reduce surface tension and can cause the breaking up of the spray stream and drops into smaller sizes, it is widely believed that adding surfactant can change the spray drop pattern by decreasing the drop size and increasing the number of drops in a given volume of water [16].

Woffinden *et al.* found that the use of surfactant decreased drop diameter in wet scrubbers by 30% [19]. Ellis *et al.* suggested that adding surfactants (Fatty alcohol ethoxylates, Polyoxyethylene sorbitan fatty acid ester, Alkoxylated ethylenediamine, or Alkyl polyglycoside at 0.5%) can reduce the volume median diameter of spray drops by up to 17%, although surface tension is not the only determinant of droplet size with surfactant solutions [20]. However, Tien and Kim have concluded that adding surfactant (Surfynol 440 and Plurafac RA 43) and increasing surfactant concentration does not change the drop size [11]. The change of spray drop pattern is related to multiple factors, in addition to spray nozzle characteristics, such as nozzle type and pressure. Other surface properties may also be important to the spray drop pattern [11, 20].

Decreasing Surface and Interfacial Tension and Increasing Coal Dust Wettability

McCully *et al.* reported that less wettable airborne particles are less likely to be removed by rain drops [21]. They have suggested that interfacial tension is important for airborne particle removal. For example, if the interfacial tension is low, the particle tends to be wetted more fully because it only needs a minimum amount of kinetic energy to successfully penetrate through drop surface tension and be engulfed. However, if the interfacial tension is high, particles need to expend their kinetic energy in

order to penetrate a drop. Otherwise, particles only adhere to the surface of the drops [21, 22, 12].

Surfactants are known to reduce the surface and interfacial tension, and therefore can allow the particle to penetrate into a drop easier. The mechanism of penetration of wetted particles into drops leads to a better loading capacity of drop collection on particles as discussed by Chander *et al.* [22, 12]. The investigators suggested that although non- or partially-wetted particles can adhere to drop surfaces and be removed by drops (as shown in Figure 1.2a), the loading capacity of drops is limited and no more particles can be collected once a particle shell is formed at the air-water interface (as shown in Figure 1.2b). However, if particles are fully wetted and the interfacial tension is low, particles can penetrate into drops, and at the same time the drop surface area also increases (as shown in Figure 1.2c and Figure 1.2d), which results in a higher loading capacity of the drop. Therefore, adding surfactant may significantly enhance the drop loading and collection effectiveness in high dust concentration situations.

Surfactants can decrease surface and interfacial tension to improve coal dust wettability. In addition, surfactants can also alter a hydrophobic state to a hydrophilic state on coal surfaces, and then form a hydrophilic wettable layer to increase surfactant absorption. This can result in better coal dust dispersion in the aqueous phase [23]. Therefore, the wetting rate by a surfactant depends upon both the properties of the surfactant and the structure of hydrophilic and hydrophobic sites on the coal particle surface, as illustrated in Figure 1.3.

Many studies have documented that various surfactants can improve the wettability of coal dust particles. Li *et al.* have suggested that anionic surfactant (0.2% sodium dodecyl sulfate) can enhance wetting performance significantly [26]. Chander has reported that nonionic surfactants (both $>3 \times 10^{-4}$ M Triton X-100 and $>1 \times 10^{-4}$ M Triton N-101) can be used as effective wetting agents on coal dust, but the choice

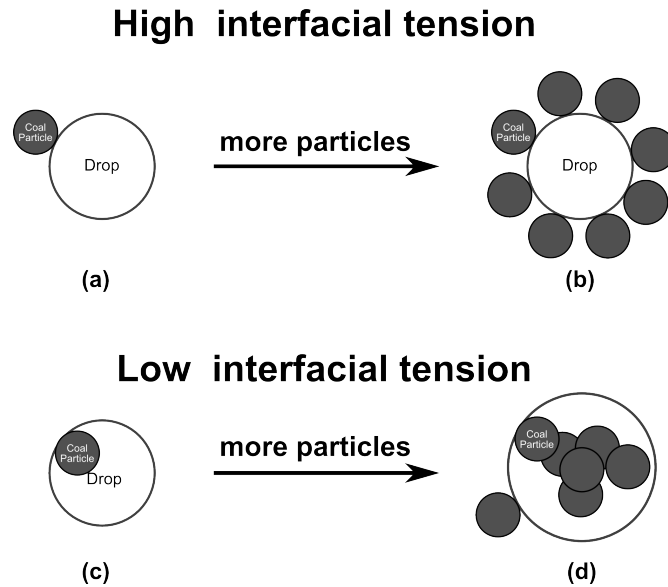


Figure 1.2: Loading capacity of a drop on collecting coal particles with high interfacial tension (a and b) and with low interfacial tension (c and d) [22, 12].

of the surfactant type should be based on the type of coal [27]. Tien and Kim also have concluded that nonionic surfactants may have better wetting qualities on coal dust because they have balanced structure consisting of hydrophilic head groups and hydrophobic tail groups [11]. They have pointed out that the wetting performances can be improved up to 215% by using sodium hydroxide and potassium hydroxide .

Altering Spray Drop Charge Characterization

When surfactant molecules dissolve in water, they orient themselves at the surface of the water. Their polar hydrophilic heads interact with water, but their non-polar hydrophobic tails tend to be repelled out of the water surface. Because the hydrophilic head of a surfactant is interacting with the water drop, its charge group can contribute to the net surface charge of a surfactant-containing drop. Therefore, when an

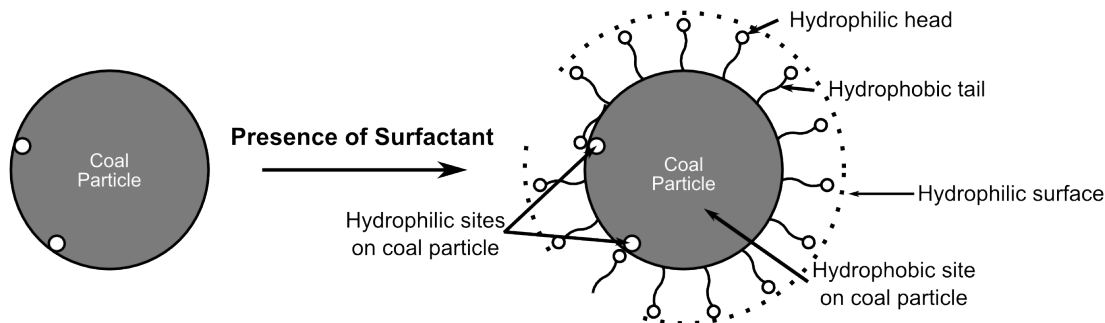


Figure 1.3: The formation of hydrophilic surface on coal particles [11, 24, 25].

anionic surfactant (e.g., sodium dodecyl sulfate (SDS)) dissolves in water, the negatively charged sulfate (SO_4^-) group may cause a net negative charge on a drop's surface. When a cationic surfactant (e.g., dodecylamine hydrochloride (DAH)) dissolves in water, the positively charged ammonium (NH_3^+) group may cause a net positive charge on a drop's surface. Nonionic surfactant (e.g., Triton X-100) has a highly polar ethylene oxide group which forms a positive dipole with CH_2 and may cause a net positive charge on a drop's surface.

Although, in theory, adding surfactant can add charges onto spray drops, studies based on electrical properties of surfactant drops were not available until the last decade. Polat *et al.* and Chein *et al.* observed that surfactant-containing spray drops can carry significant numbers of electric charges, and the sign and magnitude of charge on spray drops can vary according to the classification and concentration of surfactant applied [28, 29, 30]. According to Polat *et al.*, the fraction of positively-charged, negatively-charged, and neutral drops of distilled water were 67%, 12%, and 21%, respectively [28]. Distilled water had an average absolute value of 5,400 (either positive or negative) charges per drop for drops between 50 and 125 μm . Adding

anionic surfactant SDS to distilled water could increase the fraction of negatively-charged drops up to 77% and increase the absolute charge value to almost 20,000 (either positive or negative) charges per drop. However, SDS only increased the negative surface charges at low concentration with the peak charge at the concentration between 10^{-6} and 10^{-5} M. Once the concentration reached 3.5×10^{-3} M, the drop charge decreased to levels as low as distilled water. Adding cationic surfactant DAH could increase the fraction of positively-charged drops up to 98% at the concentration of 10^{-2} M. However, the highest absolute charge value of more than 25,000 charges per drop was observed at concentration between 10^{-6} to 10^{-5} M. In general, as the ionic surfactant concentration increased from 10^{-6} to 10^{-1} M, the charge value of ionic surfactant drop first increased to a maximum, and then decreased to a minimum when the ionic surfactant concentration was close to the critical micelle concentration (CMC). After that the charge value of ionic surfactant drop became independent with the ionic surfactant concentration [28]. Polat *et al.*, therefore, concluded that higher ionic surfactant concentration could lead to a decrease rather than a further increase in the charge value of the drop [28]. The charging behavior of the nonionic surfactant Triton X-100 was different than ionic surfactants in that it could increase the fraction of positively-charged drops to up to 95% or more and increase the absolute charge value to more than 12,000 charges per drop with a concentration of approximately 10^{-1} M. There was no CMC-related charging behavior found among nonionic surfactants [28]. Similar results were also observed by Chein *et al.*: ionic surfactants with a concentration 10-100 times lower than their CMC gave the maximum charge value on drops [30].

Zeller summarized some early studies and suggested most coal surfaces tend to carry a net negative charge. However, coal surfaces can also contain a mixture of positive and negative surface sites due to the heterogeneous composition of coal [25].

Later studies have shown that airborne coal dust after pulverization can carry different signs and magnitudes of charges according to moisture content, ambient humidity, dust components (e.g., mineral level, sulfate level), process method, and particle size [31, 32]. Therefore, the charge properties of coal dust vary depending on the characteristics of individual coal mines. Typical charge levels on respirable coal dust particles are on the order of 10 to 100 elemental charges per particle, depending on particle diameter [32], and may carry up to the order of 10^6 charges per particle for 550 μm large particles in certain condition [33].

Considering that both dust particles and surfactant-containing spray drops carry considerable amounts of electric charge, and that charged water drops can increase dust removal [34], some studies have pointed out that surfactants with the right electrical properties could potentially enhance charged coal dust collection efficiency [35, 36, 37]. For example, as early as the 1950s, Walker *et al.* suggested that the electrical attraction of dust and spray drops could lead to a dust abatement [35]. Hu *et al.* found that high drop charge level was responsible for high dust collection efficiency when using ethoxylated coco amine, a cationic surfactant [36]. In addition, Page and Organiscak pointed out that the unique dust charge properties of a coal may allow for the the addition of a proper surfactant to spray water to increase collection efficiency [37]. Because high charge levels of water drops and particles increases removal efficiency, especially for oppositely-charged water drops and particles [38], using certain types and concentrations of surfactant with high charge levels may maximize charged dust suppression.

1.3.2 Uncertainty in Surfactant Effectiveness

Even though the above mechanisms all support using surfactant in water spray to improve dust collection, the studies conducted in mines have shown mixed results. The

improvement in respirable dust control efficiency while using surfactant-containing sprays compared to plain water has ranged from zero[12] to 25%-30% [39] to more than 40% [40], and even up to 60% [41] in different studies. Surfactant seems to improve dust reduction in some mines, but not in others, and this may be caused by the use of different surfactants in different studies [23]. Therefore, a systematic study based on the classification and concentration of surfactant mainly according to electrical properties and including some of the mechanisms mentioned above, could be useful.

1.4 Objectives and Scope of the Study

The goal of this research is to reduce the respirable coal dust concentration in underground coal mines. In order to achieve the goal, the main objectives are to measure how efficiently sprays containing different classifications and concentrations of surfactants collect particles with various charge levels, and to identify factors that impact the efficiency.

To achieve these objectives, the specific aims are:

1. Measure the spray collection efficiency for monodisperse polystyrene latex spheres with different charges using several spray surfactants.
2. Measure the spray collection efficiency for polydisperse coal dust particles with different charges using several spray surfactants.
3. Design and build a test apparatus to measure the spray drop charge distribution for all tested spray surfactant solutions.
4. Analyze factors, including particle size, particle charge, drop charge, drop size, and spray surface tension, that may impact spray collection efficiency.

This dissertation consists of six chapters including the current introductory chapter. The next four chapters each contain a separate aim and are followed by the last chapter containing overall conclusions and recommendations for mining operations and future research. A bibliography and appendices are attached at the end.

Chapter 2

Factors Influencing the Control of Respirable Charged Polystyrene Latex Particles by Surfactant Sprays

Tessum, M.W., P.C. Raynor, and L. Keating-Klika. Factors influencing the airborne capture of respirable charged particles by surfactants in water sprays. *J. Occup. Environ. Hyg.* 11(9): 571-82 (2014).

2.1 Summary

This research measured the effects of particle diameter, surfactant-containing spray solution, 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 were generated in a wind tunnel. Particles were given either a neutralized, unneutralized, net positive, or net negative charge, and then were captured as they passed through sprays containing anionic, cationic, or nonionic surfactant. The remaining

particles were sampled, charge-separated, and counted with the sprays on and off at varying voltage levels to assess collection efficiency. Overall efficiencies were measured for particles with all charge levels, as well as efficiencies for particles with specific charge levels. The overall collection efficiency significantly increased with increasing particle diameter. Collection efficiencies of $21.5\% \pm 9.0\%$, $58.8\% \pm 12.5\%$, and $86.6\% \pm 43.5\%$ (Mean \pm SD) were observed for particles 0.6, 1.0, and 2.1 μm in diameter, respectively. The combination of surfactant classification and concentration also significantly affected both overall spray collection efficiency and collection efficiency for particles with specific charge levels. Ionic surfactant-containing sprays had the best performance for charged particles with opposite sign of charge but the worst performance for charged particles with same sign of charge, while nonionic surfactant-containing spray efficiently removed particles carrying relatively few charges. Particle charge level impacted the spray collection efficiency. Highly-charged particles were removed more efficiently than weakly-charged particles.

2.2 Introduction

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 Mine Safety and Health Administration (MSHA) permissible exposure limit (PEL) of 2 mg/m^3 [42, 43, 44, 45]. Even though the 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 [10]. 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 [46], 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 [23]. 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 [47]. 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 [14]. Increasing spray collection efficiency may lower dust concentrations in mines and decrease the resulting health effects.

Surfactants in the sprays are theorized to improve dust control mainly by increasing the wetting of the coal surface to prevent coal dust from becoming airborne [48, 11]. The majority of coal dust particles generated during coal cutting stay attached to the coal surface [49]. Because more dust stays attached to wetted coal surfaces than to dry surfaces [47], 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 [50, 39, 22]. In general they have found that surfactants, which have a hydrophilic head and hydrophobic tail (Figure

2.1), can 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 [51]. 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 [12] to 25%-30% [39] to more than 40% [40], and even up to 60% [41] 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, 35].

The surfactant effectiveness studies mentioned previously, conducted in the 1980s and 1990s, only considered the hydrophobic nature of the coal and surfactant. Electrical effects caused by 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.* [35] 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 [31, 37], 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 [28, 29]. Furthermore, the sign and the amount of charge varies by the classification (Figure 2.1) 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 [29].

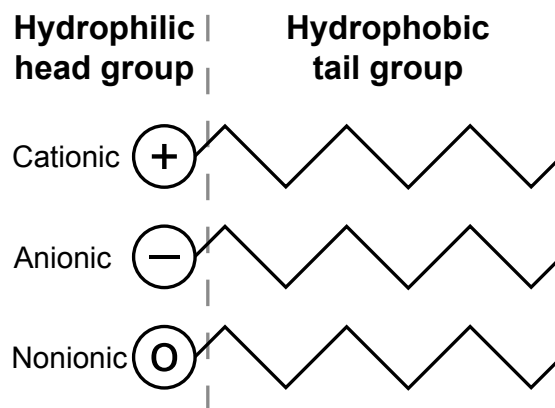


Figure 2.1: Surfactant classification based on charge present in its hydrophilic head.

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 [37]. 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.

2.3 Methods

The apparatus for this investigation had three test sections attached to a wind tunnel: (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. A schematic diagram of the apparatus is shown in Figure 2.2.

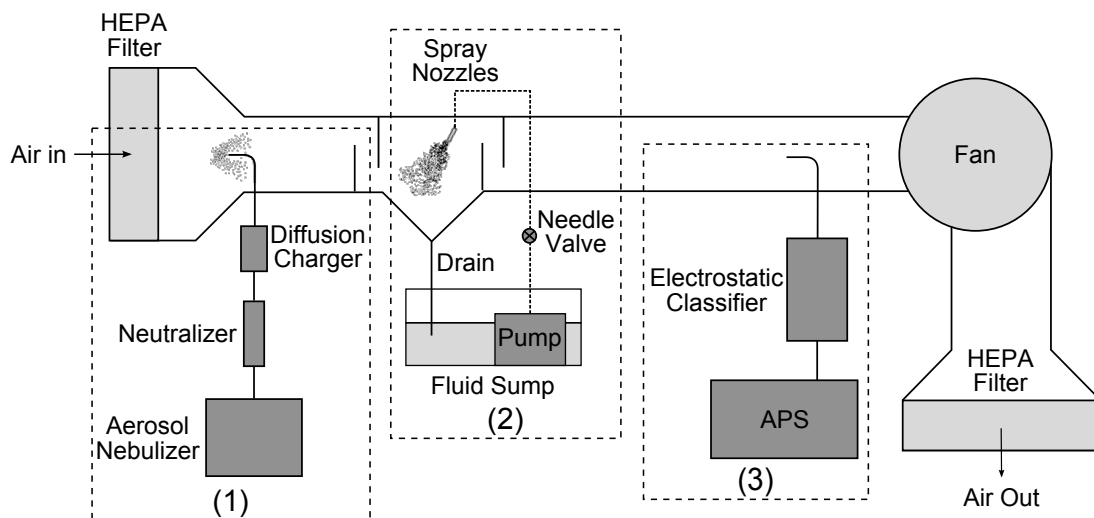


Figure 2.2: Schematic diagram of three apparatus sections: (1) the particle generation section, (2) the spray section, and (3) the particle measurement section, set in a wind tunnel.

The wind tunnel used in this study was made from acrylic plastic with a cross section of 30 cm × 30 cm. Air was drawn through the wind tunnel by a fan at a velocity of 0.61 m/s. Air entered the wind tunnel 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, polystyrene latex (PSL) spheres with nominal diameters of 0.6, 1.0 and 2.1 μm (5060A, 5100A and 5200A, Thermo Scientific, Waltham, MA) were made into separate suspensions with a concentration of 0.4% by volume using deionized and filtered water, and aerosolized using a nebulizer (TSI Inc., Shoreview, MN) 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. Preliminary tests also demonstrated that minimal air-flow through the wind tunnel, high PSL concentration in nebulizer suspensions, and high air pressure to the nebulizer produced the highest particle concentrations. Furthermore, the tests indicated that generated particle levels were steady throughout multi-hour tests, with a coefficient of variation of about 2%.

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 applied by an Eisco 5kV regulated DC power supply (Eisco Labs, Ambala Cantt, India). In this charger, the aerosol flow mixed with the ions generated by a high voltage applied corona needle, or 4. *Positive*, which was identical to negative charge condition but with the diffusion charger operating at a +5.3 kV voltage. 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. 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, with 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 [18] 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, 4.5 mm in diameter, pointed into the flow. The probe was designed according to specifications by Brockmann *et al.* [52] 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.* [53] 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 PSL 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

$$n = \frac{3Z_p\pi\mu D_p}{eC} \quad (2.1)$$

where Z_p is electrical mobility, μ is gas viscosity, D_p is particle diameter, e is elementary charge, and C is Cunningham slip correction [54].

Table 2.1: Four Measurements for Efficiency Calculation

Test Setup		Measurement	
Particle generation	Spray		
Off	Off	$B_{SprayOff}$	background particles
On	Off	$B_{SprayOff} + P_{SprayOff}$	background particles and PSL particles combined
On	On	$B_{SprayOn} + P_{SprayOn} + D$	background particles and PSL particles that are not removed by spray, plus spray droplets
Off	On	$B_{SprayOn} + D$	background particles that are not removed by spray, plus spray droplets

To measure collection efficiency, particles were counted both with and without the spray applied, as shown in Table 2.1. 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 PSL 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 PSL particles and background particles that were not removed by the spray. Collection efficiency, η , is then calculated as

$$\eta = 1 - \frac{C_{SprayOn}}{C_{SprayOff}} = 1 - \frac{(B_{SprayOn} + P_{SprayOn} + D) - (B_{SprayOn} + D)}{(B_{SprayOff} + P_{SprayOff}) - B_{SprayOff}} \quad (2.2)$$

Table 2.2: 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 dodecyl sulfate)
	Nonionic (Triton X-100)
	Cationic (Dodecylamine hydrochloride)
Surfactant Concentration	0 (Plain water)
	1×10^{-6} M (Low conc.)
	1×10^{-4} M (High conc.)

Two collection efficiencies were investigated. The overall collection efficiency, $\eta_{overall}$, for particles of all charge levels was measured when sampled particles were directly routed to the APS without passing through the electrostatic classifier. The influence on $\eta_{overall}$ of particle diameter, aerosol charge condition, surfactant classification and concentration was measured. Controlled conditions for each independent variable are given in Table 2.2. 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 to overall collection efficiency measurements, collection efficiencies for particles with specific charge levels, η_{charge} , were investigated by sampling 0.6 and 1 μm diameter particles with the classifier at 8 specific voltages and counting them with the APS. The influence on η_{charge} of aerosol charge condition and surfactant classification and concentration was also evaluated for a total of 448 combinations of test conditions, each measured three separate times.

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 analysis of variance. Scheffé's method [55] 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 [56].

2.4 Results

2.4.1 Overall Collection Efficiency

Overall collection efficiency 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 2.3. The overall collection efficiency was $21.5\% \pm 9.0\%$ (mean \pm SD) for $0.6 \mu\text{m}$ PSL particles, $58.8\% \pm 12.5\%$ for $1.0 \mu\text{m}$ particles, and $86.6\% \pm 43.5\%$ for $2.1 \mu\text{m}$ particles across all types of surfactant-containing sprays regardless of aerosol charge condition, suggesting that the effects of aerosol charge condition, surfactant classification, and surfactant concentration on overall collection efficiency may be different for each particle diameter. Because few negatively and positively charged $2.1 \mu\text{m}$ particles were observed, collection efficiencies are only reported for negatively and positively charged 0.6 and $1.0 \mu\text{m}$ particles in Figure 2.3.

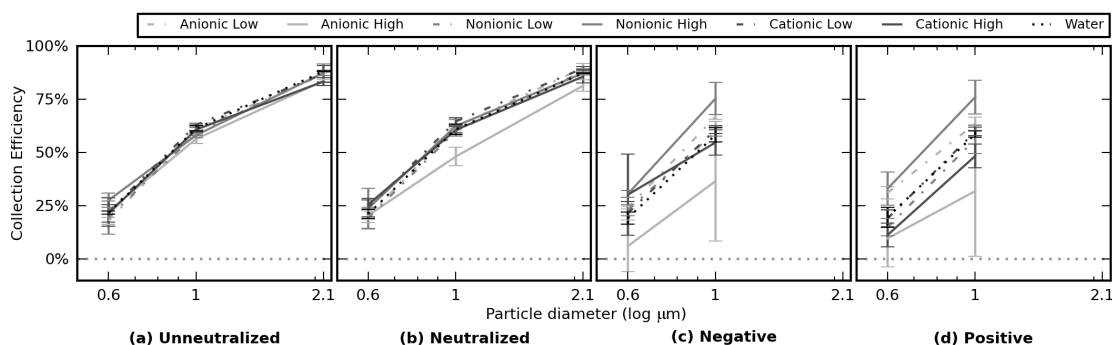


Figure 2.3: Overall collection efficiency (mean \pm SD) as a function of particle diameter for four particle charge conditions. Collection efficiencies for $2.1 \mu\text{m}$ negatively and positively charged particles are not shown owing to a large variance in results caused by a small number of total generated particles.

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 $2.1 \mu\text{m}$ particle diameters ($p = 0.78$, $p = 0.67$, and $p = 0.53$, respectively), as shown in Figure 2.4.

Spray solution type significantly affected the overall collection efficiency for both 0.6 and $1.0 \mu\text{m}$ particles ($p = 0.0087$ and $p < 0.0001$, respectively) regardless of the aerosol

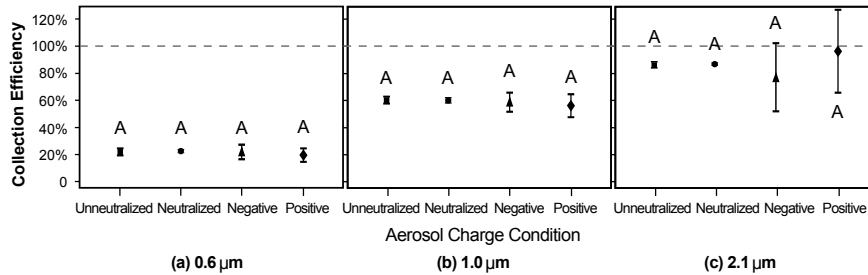


Figure 2.4: Mean overall collection efficiency with 95% confidence intervals as a function of aerosol charge condition for three particle diameters. Means with different letters are significantly different (Scheffé test, $p < 0.05$).

charge condition (Figure 2.5), 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.

Table 2.3: Comparison of Mean Overall Collection Efficiency by Surfactant Classification and Particle Diameter

Surfactant Classification	Particle Diameter (μm)								
	0.6			1			2.1		
	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%

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 2.3. 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 sprays and the plain water spray had similar efficiency for neutralized and unneutralized aerosols. Results for 0.6 μm particles are shown in Figure 2.6, and the results for 1.0 and 2.1 μm particles exhibit similar patterns.

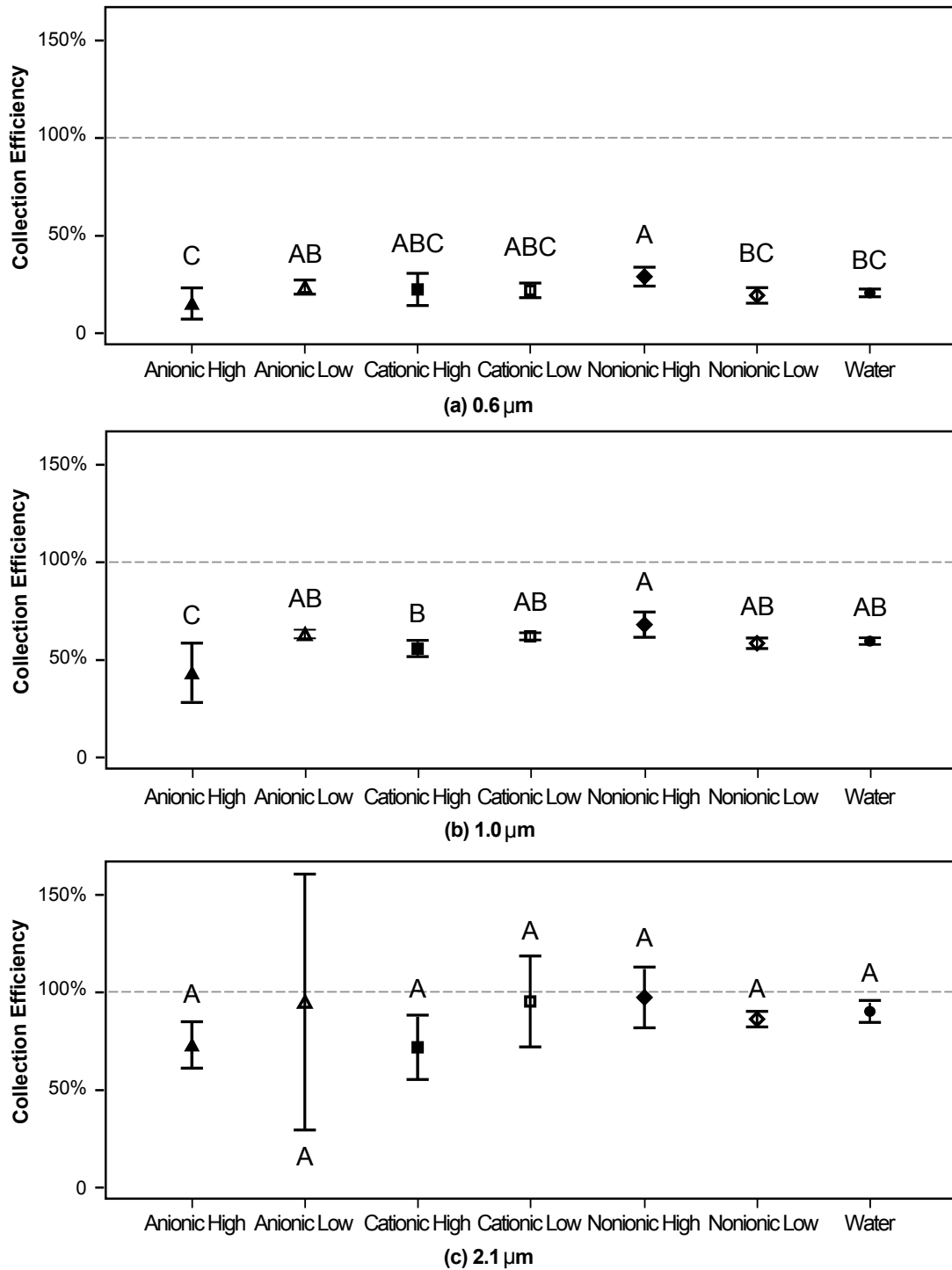


Figure 2.5: Mean overall collection efficiency with 95% confidence intervals as a function of spray solution type for three particle diameters. Means with different letters are significantly different (Scheffé test, $p < 0.05$).

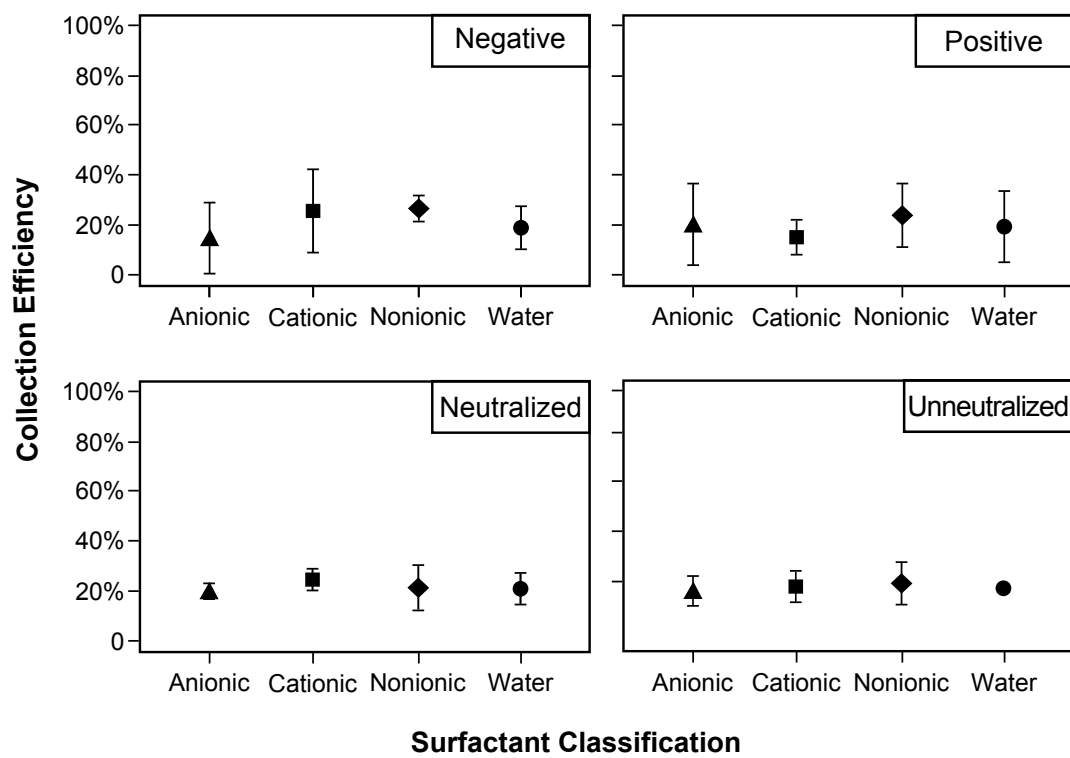


Figure 2.6: Mean overall collection efficiency with 95% confidence intervals as a function of surfactant classification on 0.6 μm particle for four aerosol charge conditions.

Table 2.4: Comparison of Mean Overall Collection Efficiency by Surfactant Classification, Concentration and Particle Diameter

Surfactant Classification	Surfactant Concentration	Particle Diameter (μm)											
		0.6				1				2.1			
		N	Mean	SD	p-value	N	Mean	SD	p-value	N	Mean	SD	p-value
Anionic	0	12	20.5%	3.3%		12	59.8%	2.8%		12	89.9%	8.6%	
	Low	12	23.4%	5.7%	0.014*	12	63.3%	3.2%	0.0047*	12	94.5%	102.5%	0.64
	High	12	14.8%	12.4%		12	43.3%	24.0%		12	72.8%	18.8%	
Nonionic	0	12	20.5%	3.3%		12	59.8%	2.8%		12	89.9%	8.6%	
	Low	12	19.2%	6.3%	0.00038*	12	58.8%	4.2%	0.00039*	12	85.9%	6.3%	0.18
	High	12	28.8%	7.5%		12	68.0%	10.2%		12	97.0%	23.8%	
Cationic	0	12	20.5%	3.3%		12	59.8%	2.8%		12	89.9%	8.6%	
	Low	12	21.8%	5.7%	0.87	12	62.1%	2.6%	0.0023*	12	94.9%	36.8%	0.11
	High	12	22.2%	12.9%		12	56.1%	6.9%		12	71.2%	25.8%	

*p<0.05 (Significant difference in overall collection efficiency)

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 2.4. 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.

2.4.2 Charge-Specific Collection Efficiency

Figure 2.7 shows the particle collection for three classifications of high concentration surfactant-containing sprays for 1.0 μm 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, as shown in Figure 2.4.

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

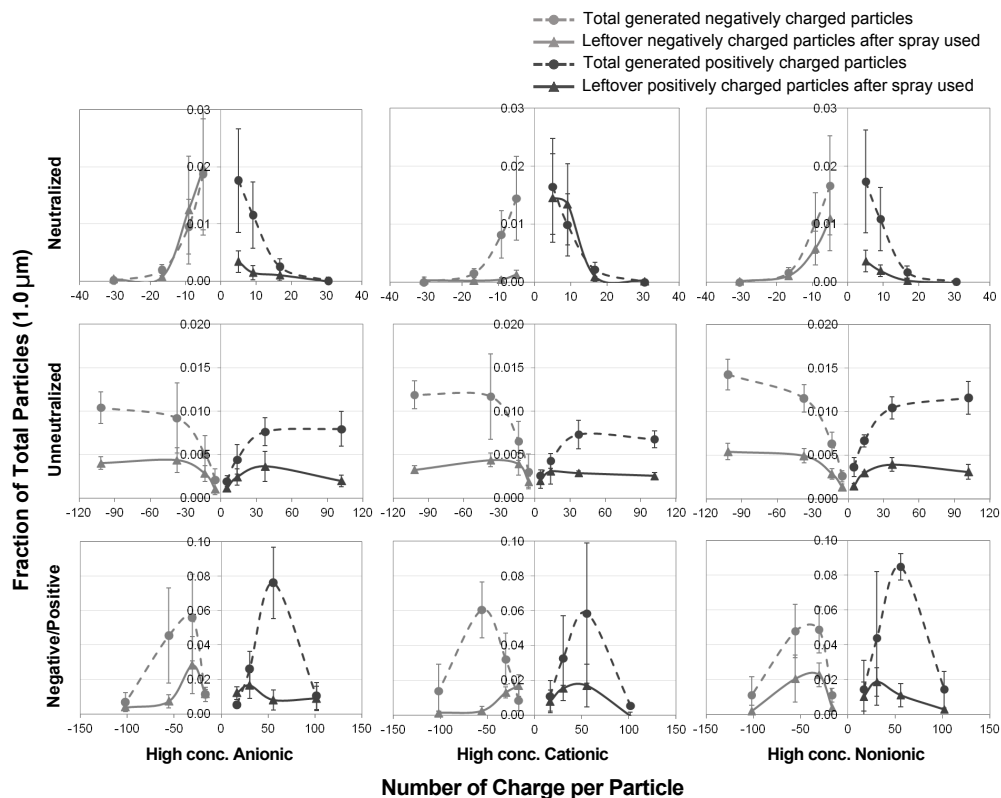


Figure 2.7: Fraction of total PSL particles (mean \pm SD) generated when spray is off and when spray is on by three classifications of surfactant spray for 1.0 μm particle for each aerosol charge condition. Neutralized particles are shown in the first row, unneutralized particles are shown in the second row, negatively (left) and positively (right) charged particles were combined in the third row.

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 opposite charged particles. For the unneutralized and positively or negatively charged aerosol, 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 2.8 and 2.9. 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 7 types of spray surfactant solutions, 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.

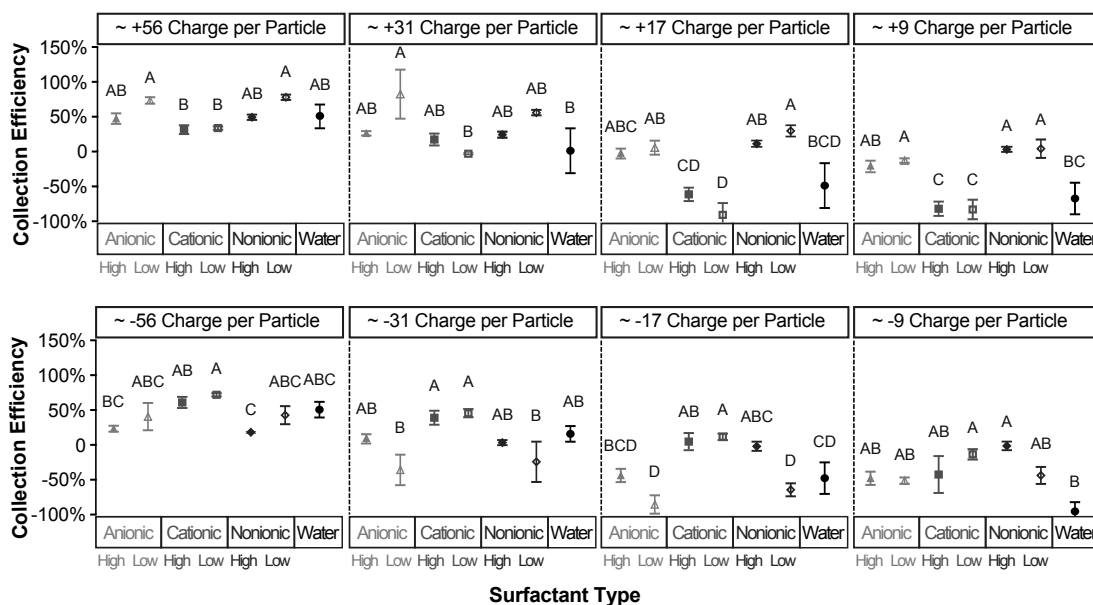


Figure 2.8: Collection efficiency (mean \pm SD) as a function of spray solution type for different charge levels of 0.6 μm particles. Means with different letters are significantly different (Scheffé test, $p < 0.05$).

Within each particle charge level, spray solution type had a substantial effect on collection efficiency. Surfactant classification contributed to this effect more than

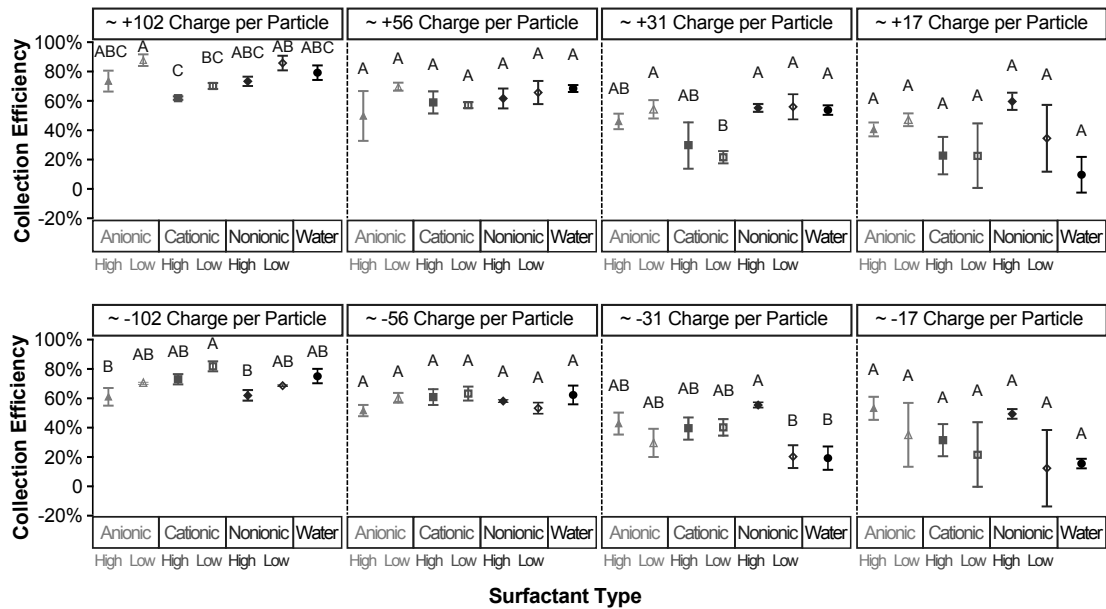


Figure 2.9: Collection efficiency (mean \pm SD) as a function of spray solution type for different charge levels of 1.0 μm particles. Means with different letters are significantly different (Scheffé test, $p < 0.05$).

did surfactant concentration, especially for 0.6 μm particles. As shown in Figures 2.8 and 2.9, Scheffé's method indicated that cationic surfactant-containing sprays had significantly higher collection 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.

2.5 Discussion

The capture of aerosol by water spray involves the physical mechanisms of inertial impaction, interception, gravity, diffusion, and electrical effects. This study investigated 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 [17], 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 2.3) regardless of test particle charge levels (Figures 2.8 and 2.9), 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 [39, 12, 40, 41] 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 2.4.

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 Walkenhorst [57], coal dust particles with a diameter of 2 μm can carry up to 300 charges per particle. 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 2.6). 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 nonionic surfactant like Triton X-100.

Certain aerosol charge conditions are more sensitive to surfactant use than others. As shown in Figure 2.7, 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 2.8 and 2.9).

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 2.8 and 2.9). 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 2.7, 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 2.2 were seen among weakly charged 0.6 μm particles (Figure 2.8). 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 $B_{\text{SprayOn}} + P_{\text{SprayOn}} + D$ to increase, which would decrease the calculated collection efficiency. This trend was consistent 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.

2.6 Conclusion

Particle size is the most important determinant of spray collection efficiency of respirable dust. Dust particles with a diameter larger than 2 μm 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.

Chapter 3

Factors Influencing the Control of Respirable Charged Coal Dust Particles by Surfactant Sprays

3.1 Summary

Surfactant-containing sprays are widely used in coal mines to control coal dust. This research investigates the effectiveness of different spray surfactants at collecting coal dust particles having various levels of electric charge. Coal dust collected from ground bituminous coal was aerosolized in a wind tunnel. The aerosol was either left unneutralized or given a neutralized, net positive, or net negative charge condition using a neutralizer and a diffusion charger. Some of the particles were removed as they passed through an anionic, cationic, or nonionic surfactant spray or a plain water spray. The remaining particles were sampled and charge-separated at different voltage levels using an electrostatic classifier. Size and concentration were then measured using an

aerodynamic particle sizer. Measurements with the spray on and off allowed calculation of efficiency. Overall collection efficiencies, including all charge levels together, were measured, as well as efficiencies of particles with specific charge levels. The tested coal dust aerosol had a geometric mean diameter of 0.89 μm and a geometric standard deviation of 1.45. The respirable mass collection efficiency was $75.5\% \pm 5.9\%$ (Mean \pm SD). Higher efficiency was observed as particle size increased: $28.8\% \pm 18.3\%$, $65.7\% \pm 9.1\%$, and $87.8\% \pm 5.7\%$ for particles 0.6, 1.0, and 2.1 μm in aerodynamic diameter, respectively. Surfactant had a significant impact on collection efficiency. In general, nonionic surfactant sprays collected as many or more of the charged particles than other sprays, especially for weakly-charged aerosols. Particle charge level significantly affected collection efficiency: strongly-charged particles tended to be collected more efficiently than weakly-charged particles. The choice of surfactant is an important determinant of the spray collection efficiency on charged coal dust particles. Nonionic surfactant may be a good choice for coal dust capture in many situations.

3.2 Introduction

Coal worker's pneumoconiosis is one of the major occupational diseases affecting coal miners, which is caused by prolonged exposure of airborne respirable coal dust. 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 [46].

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 to add in 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 [9, 22, 11, 23]. Although surfactant sprays are widely used, their ability to enhance the capture of airborne respirable coal dust varies from mine to mine [58, 41]. Although 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 [11, 23], other studies have hypothesized that electrical attraction or repulsion between dust particles and spray drops also impacts dust collection [59].

Previous studies have provided evidence that electrical effects could impact the efficiency of coal dust collection by surfactant-containing sprays. Page (2000) 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 [31]. 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 [31, 32]. Coal dust particles with a diameter of $0.5\ \mu\text{m}$ can carry on the order of 10^2 charges per particle [57], 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 [33]. However, typical charge levels on respirable coal dust particles are on the order of 10 to 100 elemental charges per particle, depending on particle diameter [32]. Electrostatic forces are most important among small particles, because for them the effects of interception and inertial impaction are negligible [60]. Although the primary particle size mode of coal dust by mass in U.S. underground coal mines is about 17-20 μm [61], 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 [29]. The sign and magnitude of charge on spray drops vary according to the classification and concentration of surfactant applied. According to Polat *et al.* [28, 29], 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.* [62] measured the efficiency with which surfactant-containing sprays collected charged, laboratory-generated, monodisperse polystyrene latex (PSL) particles with diameters of 0.6, 1.0, or 2.1 μm . They found that, in general, nonionic surfactant-containing sprays are most effective for weakly-charged respirable particles, whereas ionic surfactant-containing sprays may be 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.

3.3 Methods

The experimental set-up included particle generation, spray, and particle measurement sections in a wind tunnel, as described by Tessum *et al.* [62]. The wind tunnel has a square cross section of 0.3 m × 0.3 m. The air velocity in the wind tunnel was 0.61 m/s. The apparatus in this study differed from that of Tessum *et al.* [62] 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) was aerosolized in the particle generation section using a Model 3400 Fluidized Bed Aerosol Generator (TSI Inc., Shoreview, MN) 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 negative charge using a Kr-85 beta particle source (Model 3012A, TSI Inc.) and an optional diffusion charger (taken from a TSI Inc. AeroTrak Nanoparticle Aerosol Monitor 9000) connected to a 5kV regulated DC power supply (Eisco Labs, Ambala Cantt, India) before entering the wind tunnel. 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), cationic (Dodecylamine hydrochloride, DAH), or nonionic (Triton X-100) 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 1×10^{-4} M concentration of one of the surfactants were sprayed into the wind tunnel though three

lined hollow cone spray nozzles (No.TTD4-46, Spraying Systems Co., Wheaton, IL). 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 50 gallon 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 higher than 95% during all experiments.

In the particle measurement section, some of the remaining particles were sampled through a subisokinetic probe, optionally selected by an aerosol electrostatic classifier (Model 3071A, TSI Inc.) according to their electrical mobility, and then counted and sized by an aerodynamic particle sizer (APS, Model 3321, TSI Inc.). Overall 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 of particles with specific charge levels were measured when sampled particles were first selected by the electrostatic classifier and then counted by the APS. The APS was operated at a 60 s 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.* [62], as

$$\eta = 1 - \frac{C_{1,1} - C_{0,1}}{C_{1,0} - C_{0,0}} \quad (3.1)$$

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 Equation (3.1). 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 two-

to three-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.

Because 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 and 19.81 μm were used to calculate respirable mass concentration, C_r , as

$$C_r = \sum_{i=1}^{51} C_{n,i} \rho \frac{\pi d_i^3}{6} f_i \quad (3.2)$$

where $C_{n,i}$ is particle number concentration in size bin i ($\#/m^3$), 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 Industrial Hygienists (ACGIH)'s respirable sampling criteria [63]. Coal dust used in this study had a density of 1,310 kg/m^3 , the dynamic shape factor of coal dust is assumed as 1.05 [54]. 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.

Controlled conditions for each independent variable in overall and charge-specific collection efficiency measurements are shown in Tables 3.1 and 3.2, respectively. The factors tested in this study that may impact the overall 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 3.2 were based on the aerosol charge condition: unneutralized, neutralized, negative, or positive. Nine voltage levels were applied accordingly to each aerosol to measure the charge distribution. Because the number of

charges per particle varies with particle size, the factor of voltage level rather than number of charges per particle is shown here to avoid confusion. Efficiency measurements for each combination of these conditions were replicated three times.

Table 3.1: Test Conditions for Overall 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

Table 3.2: 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
	± 8100 V	± 8100 V	± 2430 V
	± 2970 V	± 4466 V	± 1331 V
	± 1089 V	± 2430 V	± 729 V
	± 399 V	± 1331 V	± 399 V
	0 V	0 V	0 V

The effects of aerosol charge condition and surfactant on respirable mass collection efficiency and overall number collection efficiency were estimated statistically using a two-way analysis of variance (ANOVA). Scheffé's method [55] was used to evaluate the effect of surfactant on the arithmetic means of overall collection efficiency at each aerosol charge condition. The effect of particle charge level and surfactant

on charge-specific collection efficiency was 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 and 1.0 μm in order to compare with previous PSL particle measurements. Statistical analyses were performed using R version 2.13.0 [56].

3.4 Results

3.4.1 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 Figure 3.1.

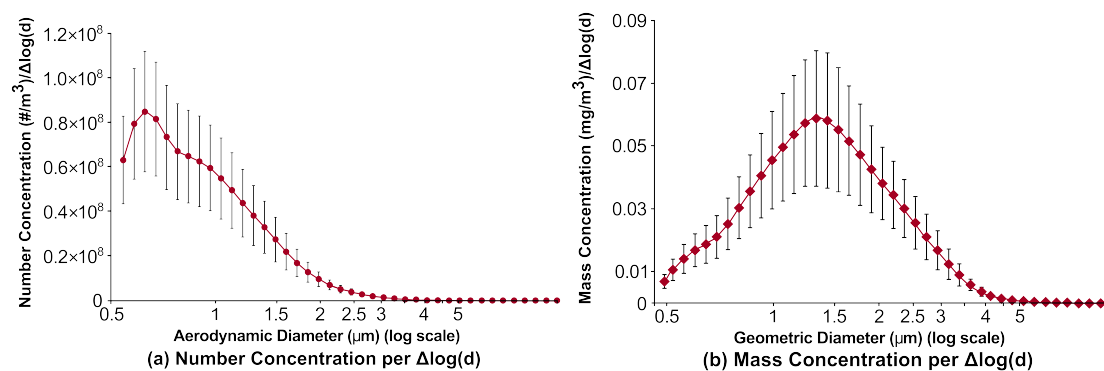


Figure 3.1: Coal dust aerosol particle concentration per $\Delta\log(d)$ (mean \pm SD, $n=12$) as a function of particle size for (a) number concentration and (b) mass concentration.

Coal dust particle charge distribution is shown in Figure 3.2, panels a-1 to a-4. Both

unneutralized and neutralized coal dust aerosols contained positively and negatively charged particles. Unneutralized aerosol had a net positive charge, while neutralized aerosol had a net negative charge. Compared to 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 did the negatively-charged aerosol, indicating that the positively-charged aerosol had higher net charge.

Figure 3.3 shows the fraction of particles carrying electric charges at eight tested charge levels for coal dust particles with 0.6 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.

3.4.2 Overall Collection Efficiency

Particle size is the most important factor affecting spray efficiency ($p < 0.0001$). Overall 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, 1.0, and 2.1 μm in aerodynamic diameter, respectively, averaged across aerosol charge conditions and surfactants. The overall number efficiency for coal dust particles for each charge condition is shown in Figure 3.4. Larger variances of overall 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 \pm 1.96SD). The average respirable mass collection efficiency for coal dust was $69.0 \pm 7.6\%$, $76.2 \pm 6.9\%$, $78.0 \pm 7.6\%$, and $78.8 \pm 7.1\%$ (mean \pm 1.96SD) for the anionic, cationic, nonionic surfactant spray and water spray, respectively. A two-way ANOVA test suggested that the 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 collection efficiency ($p = 0.875$ and $p = 0.112$, respectively).

Although the interaction of surfactant and aerosol charge condition did not significantly impact respirable mass collection efficiency, the effect of surfactant was different among different aerosol charge conditions for the overall 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 Figure 3.5. Overall, the nonionic surfactant tended to have the same or higher collection efficiency on neutralized and negatively charged aerosols relative to the other surfactants, while anionic surfactant tends to have the same or lower efficiency than the other sprays. Larger variances of overall collection efficiency are observed among negative and positive aerosol charge condition, which may mask some of the impact on overall collection efficiency by surfactants.

3.4.3 Charge-Specific Collection Efficiency

Because particle size influences the number of charges a particle can carry, as shown in Figure 3.2, 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 size levels. 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 3.3.

Comparing the leftover coal dust particles (column b in Figure 3.2) to total generated particles (column a in Figure 3.2) 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 Figure 3.6. 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.

Table 3.3: 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-value)		
Particle Size	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	<0.001*	<0.001*	<0.001*
	Unneutralized	0.874	0.314	0.0223*
1.0 μm	Positive	0.917	<0.001*	0.00355*
	Negative	0.482	0.0561	0.327
	Neutralized	0.0171*	<0.001*	<0.001*
	Unneutralized	0.466	0.0875	0.290
2.1 μm	Positive	0.141	0.0652	0.338
	Negative	0.329	0.865	0.0399*
	Neutralized	0.00382*	<0.001*	<0.001*
	Unneutralized	0.973	0.563	0.786

* p<0.05

3.5 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 larger than 2.1 μm were affected much less by charge effects (Figure 3.6). It is worth noting that particles with high charge also cause more severe health effects: Melandri *et al.* [64] 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 [62]. Comparing the charge distribution of coal dust and PSL particles at each charge condition (Figure 3.3), we found

that PSL aerosol had a larger fraction of charged particles at each charge level for 0.6 μm particles than for 1 μm particles, while coal dust aerosols had a larger fraction of charged particles for 1.0 μm particles than for 0.6 μm particles. In general, compared to 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 μm 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 μm particles. These differences were statistically significant, indicating that coal dust tends to have weaker charge properties in both quantity and magnitude for smaller particles. Because more than half of measured coal dust particles were smaller than 0.8 μm (Figure 3.1a), 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 [62]. However, ionic surfactant-containing sprays did not show significant enhancement in charged coal dust particle capture in this study. The only significant impact of ionic surfactant use is that anionic surfactant-containing spray had similar or lower efficiency on negatively charged coal dust particles compared to other surfactants. This may be because, compared to 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 [32]. In this study, there are several possible causes for the fewer particle charges observed in positively- and negatively-charged coal dust aerosol as compared to PSL. First, compared to the PSL particles, the coal dust particles went through diffusion charger at a higher concentration, which may have caused each particle to gain fewer ions. Second, the flow rate of coal dust aerosol entering the diffusion charger was about 4 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.* [32] 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 more than 300 charges per particle in the Russian study, which is much higher than our laboratory-generated coal dust particles. Because there was no particle size information given by Johnston *et al.* [32], it is not possible to make a robust charge comparison. Nonetheless, coal dust particles in underground coal mines are larger in diameter and are likely more highly charged on average than the particles generated for this study.

Undoubtedly, surfactant use affects spray efficiency. Although surfactants did not significantly impact overall collection efficiency for all aerosol charge conditions, non-ionic surfactant sprays had better overall performance on weakly charged neutralized aerosols and on negatively-charged aerosols, which had fewer charges per particle

compared to the unneutralized and positively-charged aerosols (Figure 3.2). The observation that nonionic surfactant can significantly remove weakly charged particles concurs with previous PSL results [62], 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 [11], 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 [19]. 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.* [28]: Polat *et al.* found that, for a given surfactant concentration, ionic surfactant-containing sprays carry a net corresponding charge and nonionic surfactant spray has weak positive charge. They also found that plain water has an even weaker positive charge than nonionic surfactant and that adding ionic surfactants substantially enhances the charge level of spray drops. However, differences in our experimental setup relative to that of Polat *et al.* [28] (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.* [28]. 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 $0.6\ \mu\text{m}$ particles (Figure 3.6). As discussed in

Tessum *et al.* [62], 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 Figure 3.3. 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 Equation (3.1), and resulting in the underestimate 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. First, drop evaporation may cause charging as described by Lear and Harmon [65]. 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. Second, 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 smaller than $0.6 \mu\text{m}$ could collide with a neutral drop also having a diameter smaller than $0.6 \mu\text{m}$, and the combined particle and drop could be measured by the APS as a charged $0.6 \mu\text{m}$ 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.

3.6 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 overall 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 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.

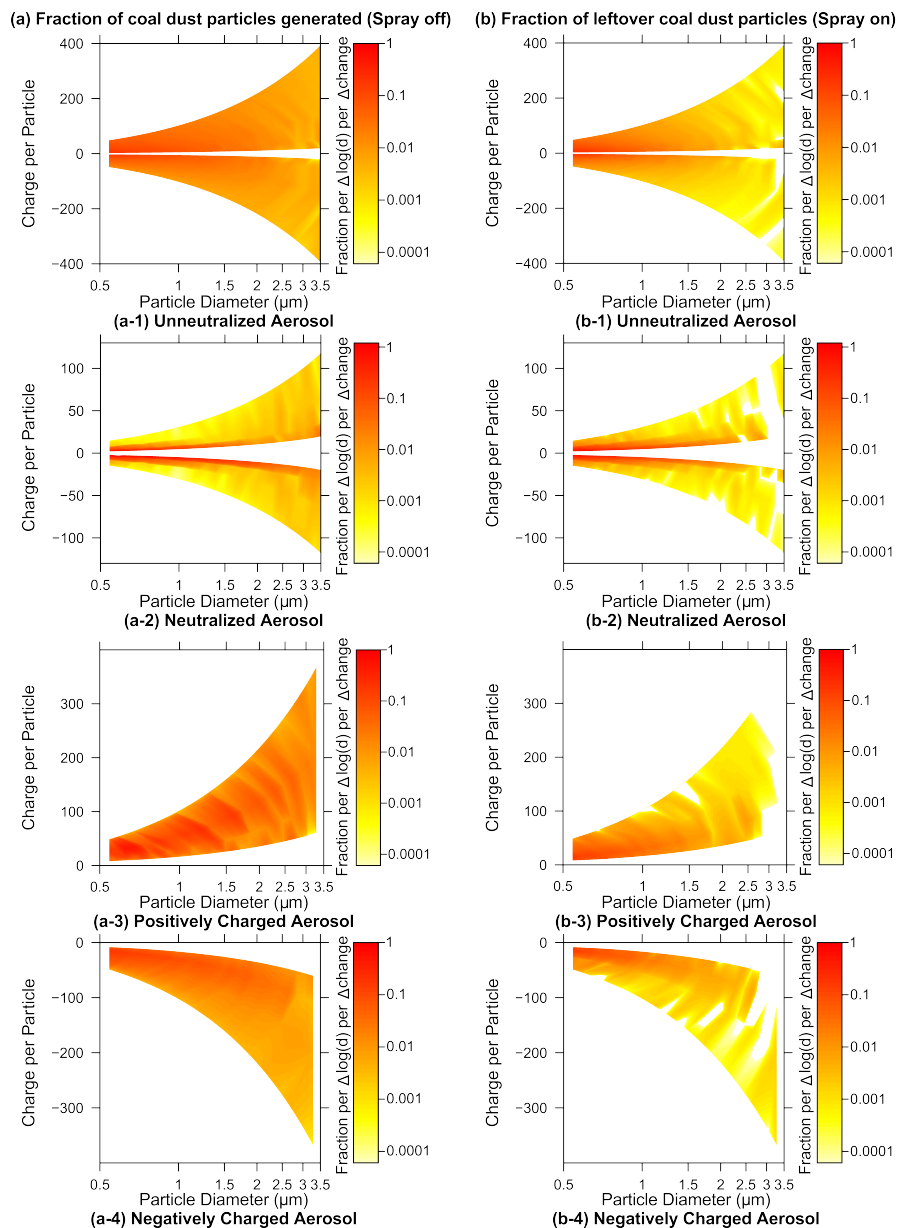


Figure 3.2: Mean fractions of charged coal dust particles at different particle sizes and charge levels when (a) spray is off and (b) when spray is on for four (1-4) aerosol charge conditions, normalized by both $\Delta \log(d)$ and $\Delta(\text{number of charges})$. Values in panels b1-4 represent averages among all three surfactants and plain water.

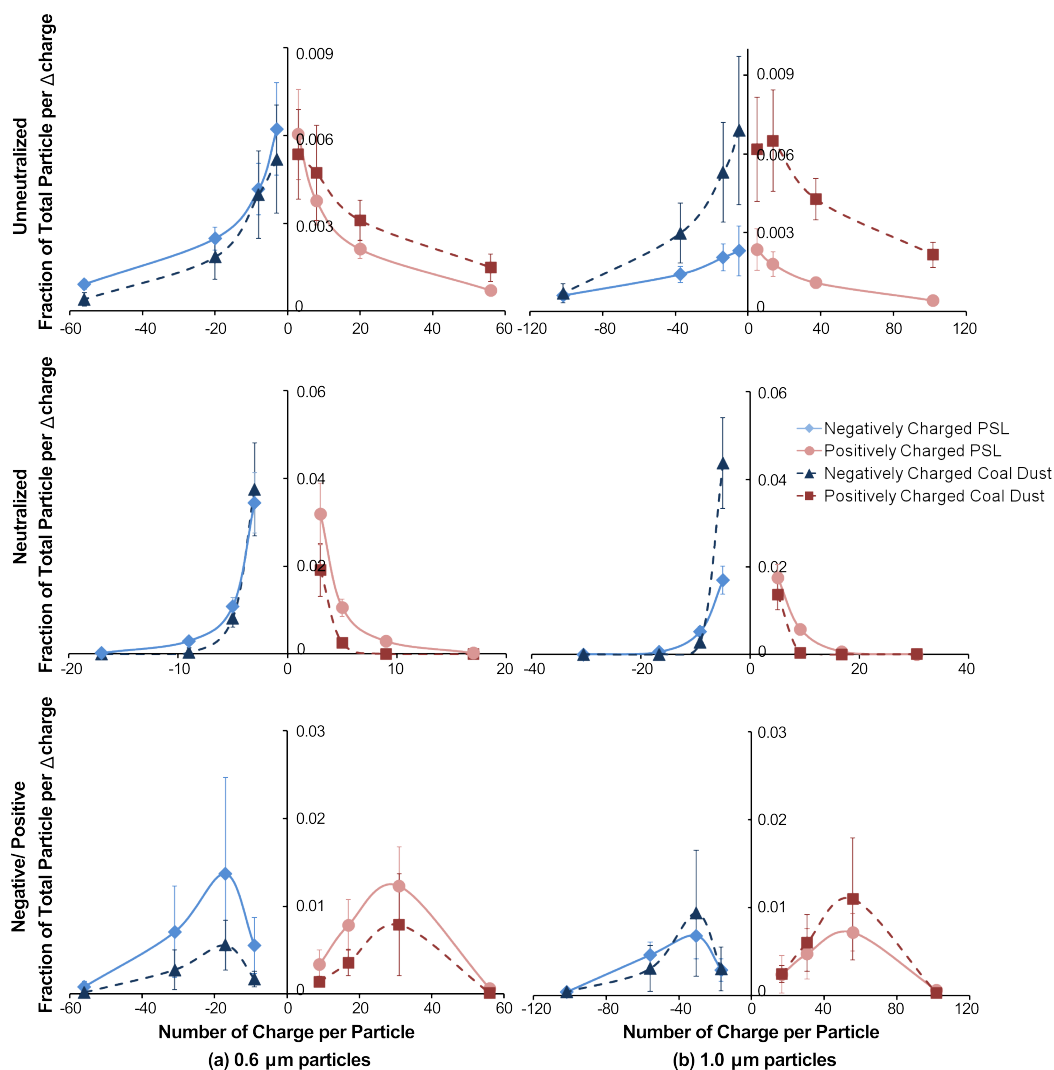


Figure 3.3: Comparison of fractions of total generated particles (mean \pm SD) of PSL and coal dust at different charge levels for (a) 0.6 μ m and (b) 1.0 μ m particle size for four aerosol charge conditions: Unneutralized particles are shown on the top row, neutralized particles are shown in the middle row, negatively (left) and positively (right) charged particles are combined in the bottom row. The fraction was calculated by the number of 0.6 or 1.0 μ m particles with a given charge divided by the total number of generated particles of the same size, and normalized by Δ (number of charges).

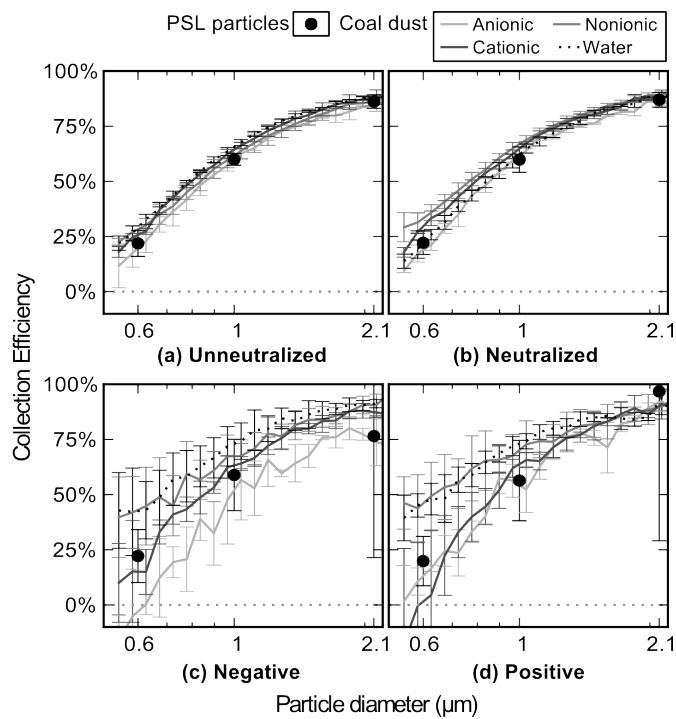


Figure 3.4: Overall collection efficiency (mean \pm SD) of coal dust and PSL particles as a function of particle diameter for four aerosol charge conditions. The overall collection efficiencies of PSL particles were averaged across different types of spray.

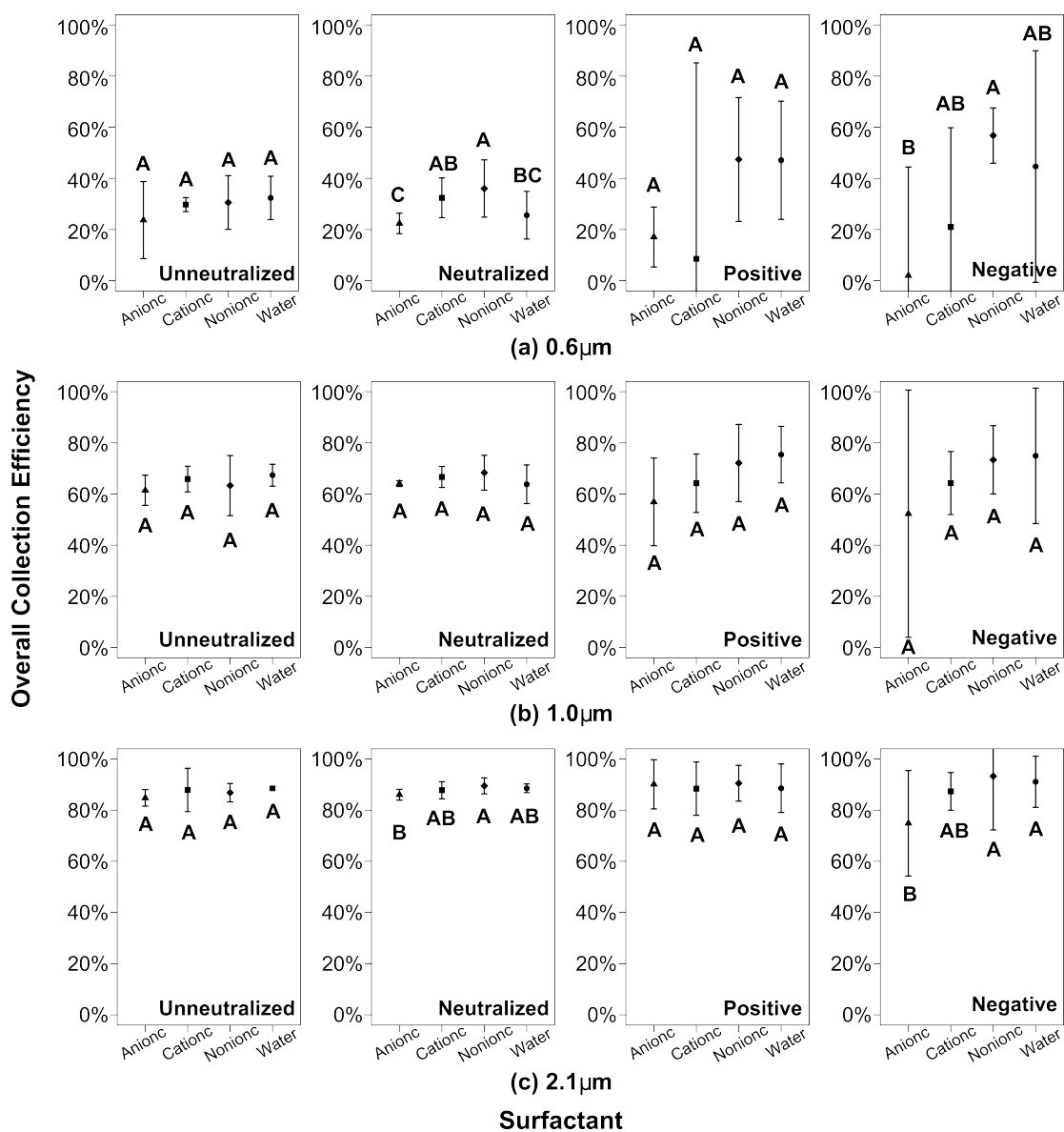


Figure 3.5: Mean overall number collection efficiency with 95% confidence intervals as a function of surfactant for four aerosol charge conditions for three particle sizes at 0.6, 1.0 and 2.1 μm . Within each figure, means with different letters are significantly different (Scheffé test, $p < 0.05$).

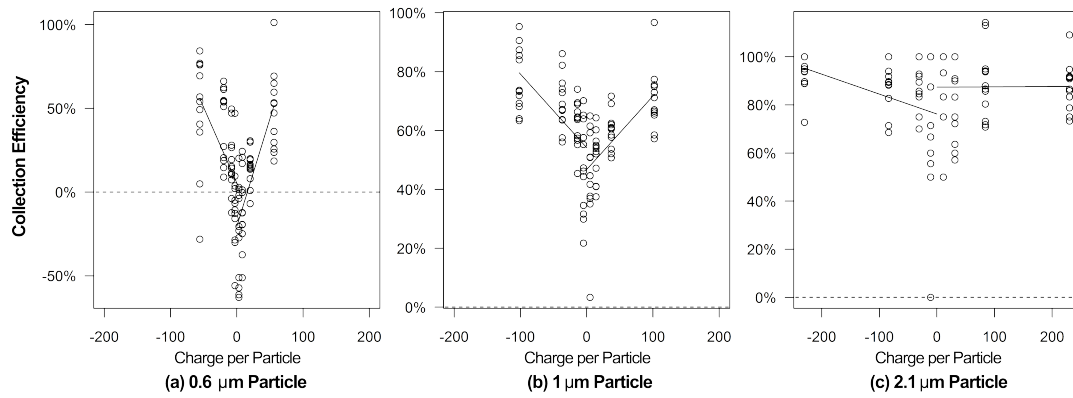


Figure 3.6: Scatter plot with regression line of collection efficiency on unneutralized coal dust aerosol as a function of particle charge level for three particle sizes.

Chapter 4

Measuring Electrostatic Charge on Spray Drops

4.1 Summary

A new spray drop charge measurement system was developed to measure a broad range of sizes and electrostatic charges for spray drops based on the principle that the free falling drops can be separated according to their electrostatic mobility in an electrical field with known strength. High and low concentration anionic, cationic, and nonionic surfactant sprays and water spray were tested. Anionic surfactant-containing sprays had the largest average drop sizes, and water spray had the smallest average drop sizes. Drop charge level was significantly higher among larger size drops. Nonionic and cationic surfactant-containing sprays and water spray carried net positive charge on average, while anionic surfactant-containing sprays tended to carry net negative charge on average. Increasing surfactant concentration did not increase the magnitude of drop charge. Although the spray drop charge measurement system developed in this study can be used to measure spray drop electrostatic charge

distribution, future design improvements should be considered in order to improve accuracy.

4.2 Introduction

The U.S. government estimated that the deaths of more than 76,000 miners nationwide between 1968 and 2010 were either directly or partially caused by coal workers' pneumoconiosis (CWP; otherwise known as black lung), which is associated with long term occupational exposure to airborne respirable coal dust [3]. Although respirable coal dust control has been a priority for many years, data from the Mine Safety and Health Administration's inspector samples indicate that concentrations of respirable coal dust in underground coal mines are still not completely in compliance with the federal occupational exposure limits. During 2004 and 2008, about 22% of coal mine respirable dust measurements across all occupations among underground coal workers were in exceedance of the 1 mg/m^3 recommended exposure limit (REL), and about 4% of coal mine respirable dust measurements exceeded the previous permissible exposure limit (PEL) of 2 mg/m^3 [8].

Water-based spray systems are one of the primary dust control measures in coal mines. However, properly designed water spray systems can only reduce respirable dust by an average of 30% under actual mining conditions [14]. Many researchers have suggested using surfactants as additives in spray water to enhance the dust suppression, but spray effectiveness studies with surfactants in mine trials have not consistently shown improvement [39, 40, 12, 41]. A better understanding of how adding surfactant impacts the capture of respirable particles by spray drops would be a key to explaining the inconsistency among previous studies, and it would guide surfactant selection for better collection efficiency.

The presence of surfactant in water-based sprays can enhance the capture of respirable particles depending on surfactant type and particle charge characteristics [62]. This observation may be caused by electrical effects between spray drops and respirable particles because highly charged particles tend to be removed more efficiently by surfactant-containing spray that theoretically carry opposite charges on average [62]. However, there is no consensus in the literature regarding the mechanism of how surfactants impact dust suppression. One hypothesis is that surfactants improve dust capture by reducing the surface tension of the water drops and changing the polarity of the dust through adsorption [23, 11]. Another hypothesis is that surfactants alter the electrical properties of spray solutions (or drops), and therefore improve the suppression of charged dust [36]. Some studies also have argued that surfactants improve the dust capture by altering the drop parameters, such as drop size [19].

Although, in theory, the electrostatic charge of the spray solution or drop depends on the classification of surfactant which is based on the polar functional groups dissolved in water, both mechanical and chemical factors impact the electrostatic charge on drops during generation. For example, the atomization of liquid in spray can generate electrostatic charges on spray drops through friction with the spray nozzle as the liquid is sprayed. During atomization, the electrostatic charge on spray drops of most liquids, including distilled water, tends to be positive [66]. The combination of surfactant classification and concentration also impacts electrostatic charge on spray drops [28]. Anionic surfactant-containing spray drops are most likely to carry negative charges, while cationic and nonionic surfactant-containing spray drops tend to be positive [29]. The role of surfactant concentration on spray charge is complicated, but in general there is no consistent correlation between surfactant concentration and drop charge level [36, 28].

In order to argue that the improvements in spray collection efficiency observed in

our previous studies is caused by the electrical effect, the qualitative and quantitative measurement of electrostatic charge on spray drops is essential. There is not a standard method to measure charge distribution on nozzle-generated spray drops owing to the wide range of possible drop diameters, which can range from several micrometers to more than 1000 μm , as well as instability in composition due to evaporation and changes in shape. Polat *et al.* [28] described a method to measure the electric charges on individual spray droplets based on the Millikan oil drop method. However, the work of Polat *et al.* [28] only measured a limited number of spray drops, and may not accurately represent the charge characteristics of the entire spray. In addition, the method of Polat *et al.* [28] did not include size information for the surfactant-containing spray drops, even though the electrostatic charge on drops varies with drop size.

In this study, a spray drop charge measurement system was developed based on the method of Polat *et al.* [28] to measure a broad range of sizes and electrostatic charges for spray drops. This new system allows the measurement of electrostatic charge distribution on surfactant-containing spray drops, and therefore helps to determine how the presence of surfactant alters the spray drop charge distribution.

4.3 Methods

4.3.1 Materials

The surfactant classifications and concentrations tested were selected among those that were evaluated by Polat *et al.* [28, 29] and used in previous collection efficiency research [62]. The surfactants were chosen according to their classification: anionic (Sodium dodecyl sulfate, Fisher Scientific Inc., NJ), nonionic (Triton X-100, Fisher Scientific Inc., NJ), and cationic (Dodecylamine hydrochloride, Tokyo Chemical Industry

Co., Japan).

The high surfactant concentration was chosen to be 1×10^{-4} M because it was the maximum concentration of surfactant-containing solution that gave a consistent spray generation [62]. The low surfactant concentration was chosen to be 1×10^{-6} M, and plain deionized water was used as a control.

4.3.2 Methods

Tests were conducted in an acrylic plastic wind tunnel with a spray section and a drop measurement section as shown in Figure 4.1. The wind tunnel was parallel to the ground with a cross section of 30 cm \times 30 cm. Clean air was drawn through the wind tunnel by a fan at a velocity of 0.61 m/s. This air velocity allowed most spray drops to fall between baffles located upstream and downstream of the spray section.

A spray solution, made using one combination of the surfactant classifications and concentrations mentioned above, was stored in a 50 gallon fluid container and then pumped into three hollow cone spray nozzles (No.TTD4-46, Spraying Systems Co., Wheaton, IL) with a flow of 2.95 L/min to each nozzle at a pressure of 552 kPa. The spray cones had an angle of 33° from vertical. According to Tien and Kim [11], the drop sizes produced by the spray nozzle were not expected to decrease substantially when the surfactant is added. In addition, the majority of spray drops had a Sauter mean diameter between approximately 90 and 210 μ m when using the same single nozzle with the same flow rate, pressure and cone angle settings [18]. In order to avoid inconsistency and to decrease the waste of surfactant solution, the surfactant solutions were drained back into the fluid container after being sprayed and were recirculated by a pump (Model 4UP51A, Dayton Electric Manufacturing Co., Niles, IL). During the recirculation, the surfactant solutions were drawn through a polypropylene filter (Model 08PT, Keystone Filter, Hatfield, PA) to prevent contamination before being

resprayed into the wind tunnel.

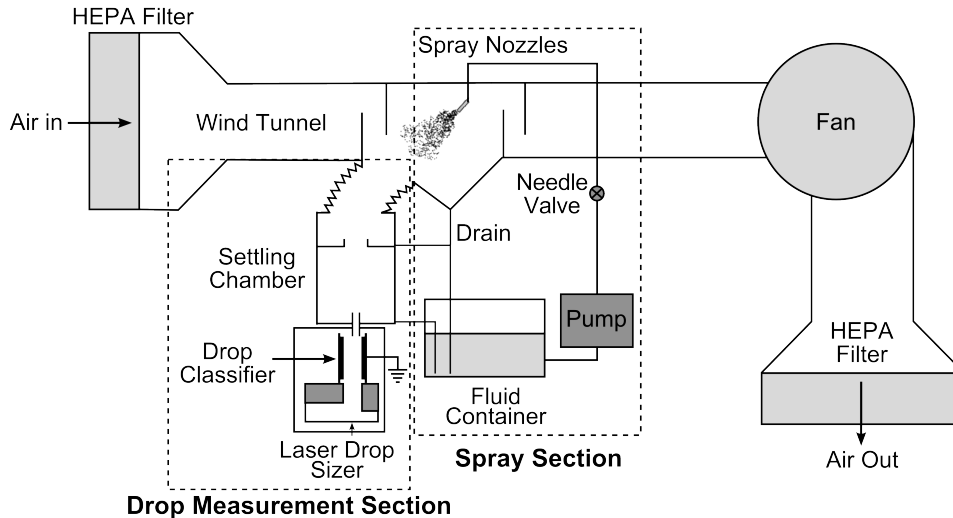


Figure 4.1: Schematic diagram of the spray section and the drop measurement section

A drop measurement section was designed based on the work of Polat *et al.* [28] as shown in Figure 4.1. During size-specific drop charge measurement, a fraction of the spray drops of each surfactant solution fell into a spray drop charge measurement system starting with a vertical cylinder settling chamber, 47 cm high, which allowed most drops to reach terminal settling velocity. The interior walls of the settling chamber were wetted with distilled water before each test to reduce static charging and to saturate the air with water vapor to minimize spray drop evaporation. At the end of the settling chamber, a grounded metal square tube with an opening size of $d_1 = 6$ mm (about 70 times as an average drop size) connected the settling chamber to a drop classifier, shown in Figure 4.2. This small cross-section drop inlet allowed drops with small deflection from the vertical path to pass through. One edge of the tube was 1 mm from a grounded electrode, and the other edge of the tube was at the center line

between the two parallel electrodes. In the drop classifier, drops were separated according to their electrostatic mobility and then sized by a laser drop sizer (Model D30, Oxford Laser Ltd. Shirley, MA) at the bottom of the drop classifier. The Model D30 laser drop sizer is a real time instrument with a minimum detectable size limit of 20 μm . Drop size measurements were recorded in fifteen bins, log-equally divided from 20 to 320 μm .

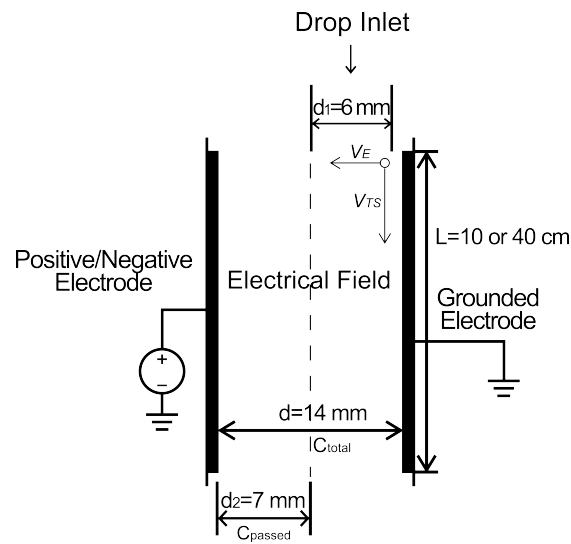


Figure 4.2: Schematic diagram of the drop classifier

The overall drop size distribution was also measured by a laser drop sizer at the top of the settling chamber for each surfactant solution that was sprayed into the wind tunnel. This information was combined with the drop charge and size information that was recorded for drops that passed through the drop classifier to yield the overall charge distribution for each spray type.

4.3.3 Theory/ Calculation

The design principle for the drop classifier was to separate the free falling drops according to their electrostatic mobility in an electrical field with known strength. The electrical field was created by two parallel vertical stainless-steel rectangular plates. One plate was grounded and the other one was charged by a DC high-voltage power supply with a maximum voltage of ± 8 kV. Drops were assumed to fall into the electrical field at terminal setting velocity, V_{TS} , calculated as

$$V_{TS} = \frac{\rho_d D_d g}{18\mu}, \text{ when } Re_p < 1 \quad (4.1)$$

$$V_{TS} = \left(\frac{4\rho_d D_d g}{3C_D \rho_g} \right)^{\frac{1}{2}}, \text{ when } Re_p > 1 \quad (4.2)$$

in which ρ_d is drop density, D_d is drop diameter, g is acceleration of particle due to gravity, μ is air viscosity, ρ_g is air density, and C_D is drag coefficient, a function of Reynolds number. $C_D = \frac{24}{Re}$ for $Re < 1$, and $C_D = \frac{24}{Re}(1 + 0.15Re^{0.687})$ for $Re > 1$.

The movement of a drop in the vertical direction was assumed to be at constant velocity V_{TS} , as shown in Figure 4.2. As they passed through the electric field, the drops also accelerated toward the terminal electrostatic velocity in the horizontal direction. The terminal electrostatic velocity, V_{TE} , was calculated as

$$V_{TE} = \left(\frac{neE}{C_D \frac{\pi}{8} \rho_g D_d^2} \right)^{\frac{1}{2}} \quad (4.3)$$

in which n is number of charges, e is elementary charge, and E is field strength. In addition to the knowledge of the spray drop size range given by Pollock and Organiscak [18], Polat *et al.* [28] indicated that water and surfactant containing drops carry on the order of 1,000 charges per drop, ranging from 0 to 15,000 charges. Therefore, the drop classifier was designed to separate the drops across a wide range of size and electrical charge. Two sets of the electrodes with lengths (L) of 10 cm and 40 cm and

with the distance between the electrodes of $d=14$ mm were applied in order to measure a wide electrostatic mobility range. However, large drops, especially drops larger than $200 \mu\text{m}$, took relatively long times to reach their terminal electrostatic velocity, and therefore the non-steady state electrostatic velocity (V_E) of drops as they passed through the electrical field was accounted for in the following manner. Drops in the drop classifier experienced drag (F_D) and electrostatic (F_E) forces in the horizontal direction. Until the drops reached the terminal electrostatic velocity, the electrostatic force was larger than the drag force, causing acceleration in the horizontal direction. The governing equation describing drop horizontal movement was

$$F_E - F_D = ma \quad (4.4)$$

in which m is drop mass, a is acceleration, electrostatic force $F_E = neE$, and drag force $F_D = \frac{C_D \pi \rho_g D_d^2 V_E^2}{8}$. The acceleration due to electrostatic forces was then calculated as

$$a = \frac{6neE}{\pi D_d^3 \rho_d} - \frac{3C_D \rho_g V_E^2}{4D_d \rho_d} \quad (4.5)$$

Traveling each small distance ΔY in the vertical direction required time $\Delta t = \frac{\Delta Y}{V_{TS}}$. The drop traveled through the electrical field in total time $t = \frac{L}{V_{TS}}$. Equation 4.5 was integrated numerically to determine instantaneous electrostatic velocity, electrostatic acceleration, and the distance the drop traveled in the x and y axial direction. The algorithm shown in Table 4.1 explains the principle of the calculation. This model of the motion of the drops was combined with measurements of drop size and fraction of drops passing through the drop classifier to estimate the number of charges on each drop as described in Section Data Inversion.

The laser drop sizer was used to count and size drops at two locations according to whether an electrical field was created or not in the drop classifier. When there was no electrical field created, the laser drop sizer measured all drops that passed through

Table 4.1: The Algorithm for Calculating Horizontal Drop Movement in Electrical Field

Timestep	Horizontal Velocity	Horizontal Acceleration	Horizontal Distance
$t_0 = 0$	$V_{E(0)} = 0$	$a_0 = \frac{6neE}{\pi D_d^3 \rho_d} - \frac{3C_D \rho_g V_{E(0)}^2}{4D_d \rho_d}$	$X_0 = 0$
$t_1 = \Delta t$	$V_{E(1)} = V_{E(0)} + a_0 \cdot \Delta t$	$a_1 = \frac{6neE}{\pi D_d^3 \rho_d} - \frac{3C_D \rho_g V_{E(1)}^2}{4D_d \rho_d}$	$X_1 = X_0 + V_{E(1)} \cdot \Delta t$
\vdots	\vdots	\vdots	\vdots
$t_i = i \cdot \Delta t$	$V_{E(i)} = V_{E(i-1)} + a_{(i-1)} \cdot \Delta t$	$a_i = \frac{6neE}{\pi D_d^3 \rho_d} - \frac{3C_D \rho_g V_{E(i)}^2}{4D_d \rho_d}$	$X_i = X_{(i-1)} + V_{E(i)} \cdot \Delta t$

the drop classifier at the bottom of the classifier across the entire width of $d = 14$ mm. When the electrical field was created, the laser drop sizer measured the drops that passed through the left half of the bottom of the drop classifier with a width of $d_2 = 7$ mm, as shown in Figure 4.2. The test system described here was only able to sample of drops with the opposite sign of charge as the charged electrode. For example, when negative voltage was applied on the charged electrode, the positively charged drops were attracted and moved toward the charged electrode, and thus some of them were sampled at d_2 located next to the charged electrode. On the other hand, the negatively charged drops were repelled and moved toward the grounded electrode, and thus were not sampled. Because of this, tests were run with both positively and negatively charged electrodes and the results were combined. Positive and negative voltages were applied to the electrode to selectively pass drops through the drop classifier, allowing a wide charge and size distribution measurement of the drops. Thirty voltage levels ($\pm 100V$, $\pm 200V$, $\pm 300V$, $\pm 400V$, $\pm 500V$, $\pm 600V$, $\pm 800V$, $\pm 1000V$, $\pm 1500V$, $\pm 2000V$, $\pm 3000V$, $\pm 4000V$, $\pm 5000V$, $\pm 6000V$, and $\pm 8000V$) were tested for both the 10 cm and 40 cm electrodes.

The horizontal distance that a drop moves through the electrical field, X , can

be calculated using the algorithm in Table 4.1. Assuming that drops are uniformly distributed as they enter the electric field, the theoretical Drop Penetration Fraction ($F_{theoretical}$) of drops passing through the drop classifier was calculated by using the algorithm to track the theoretical path of many drops that started in random locations along d_1 in Figure 4.2 as they fell through the drop classifier and observing how many of the theoretical particles also pass through d_2 . The fraction of particles that the algorithm predicted would pass through d_2 was the theoretical Drop Penetration Fraction. Figure 4.3 shows the theoretical drop sampling efficiency for the spray drop charge measurement system as a function of drop size and charge for an applied voltage of +1000 V.

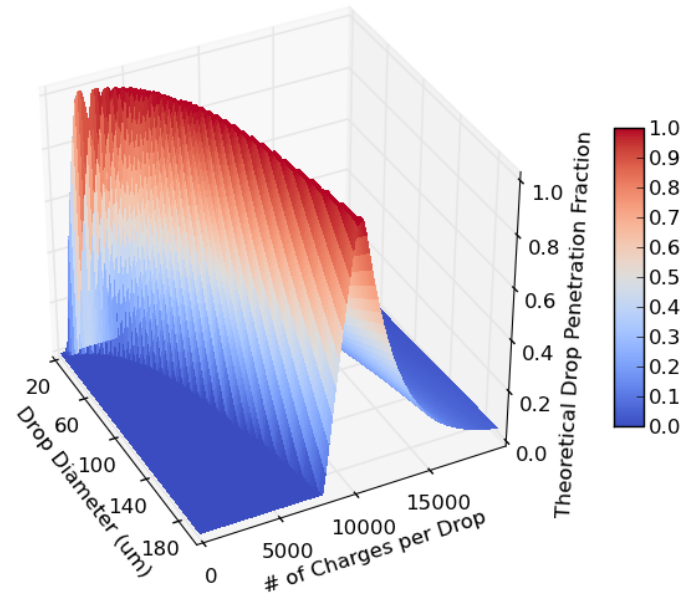


Figure 4.3: Fraction of the drops passed through the electrical field vs. drop size and charge distribution when +1000 V is applied

The measured Drop Penetration Fraction ($F_{measured}$) can also be calculated as in Equation 4.6:

$$F_{measured} = \frac{C_{pass}}{C_{total}} \quad (4.6)$$

where C_{total} is total drop count measured across the total width d when both electrodes were grounded and C_{pass} is the drop count of those passing through the drop classifier at d_2 when an electrical field was applied. Both C_{pass} and C_{total} were measured five times for each test condition. With measurements from multiple voltages, a data inversion technique was used to compare $F_{measured}$ to $F_{theoretical}$ to reconstruct the charge distribution for each drop size interval.

Once the drop charge distribution was obtained, the net average charge on spray drops was also calculated for each spray type by separately summing the positive and negative charges, q_i , with each fraction of charged drops at each drop sizes of all the sampled drops for each surfactant spray solution. The result was then divided by the total number of positively- or negatively-charged drops, $\sum_{i=1}^{30} [\sum_{i=1}^{15} n_j |D_{d,i}]$, as

$$Net\ average\ charge = \frac{\sum_{i=1}^{30} [\sum_{i=1}^{15} [q_j n_j] |D_{d,i}]}{\sum_{i=1}^{30} [\sum_{i=1}^{15} n_j |D_{d,i}]} \quad (4.7)$$

where i is one of the thirty voltages with both signs applied on drop classifier, and j is one of the fifteen drop sizes.

4.3.4 Data Inversion

In order to maintain a manageable computing workload for data analysis, drop charge distribution was analyzed with fifteen drop size intervals, log-equally divided from 20 to 320 μm , and fifteen drop charge intervals for each sign of charge log-equally divided from 1 to 150,000 charges per particle for data collected using the 10 cm electrical plates, and multiple sets of fifteen drop charge intervals for each sign of charge log-equally divided up to 1,500,000 charges per particle for data collected using the 40 cm

electrical plates.

The theoretical Drop Penetration Fraction ($F_{theoretical}$) is a function of drop size D_d , drop charge C_d , and voltage V applied on the device. At each voltage applied, the measured Drop Penetration Fraction ($F_{measured}$) for a specific D_d , represents an average of drops with different combinations of $F_{theoretical}$ as in Equations 4.8:

$$\begin{aligned}
 F_{measured}(V_1) &= \sum_{i=1}^{15} [n_i \cdot F_{theoretical}(D_d, C_{di}, V_1)] + \epsilon_1 \\
 F_{measured}(V_2) &= \sum_{i=1}^{15} [n_i \cdot F_{theoretical}(D_d, C_{di}, V_2)] + \epsilon_2 \\
 &\vdots \\
 F_{measured}(V_{15}) &= \sum_{i=1}^{15} [n_i \cdot F_{theoretical}(D_d, C_{di}, V_{15})] + \epsilon_{15}
 \end{aligned} \tag{4.8}$$

where V is one of the fifteen voltages for each sign of charge applied on drop classifier, ϵ is experiment error for each measurement, and n_i is the fraction of drops with charge C_{di} . For different voltages V applied on the device, each n_i is a function of drops with the specific drop size D_d and drop charge C_{di} at voltage V . Additionally, each n_i must take a value between 0 and 1, and the sum of all n_i must be less or equal to 1, as in Equation 4.9:

$$0 < n_i < 1, \text{ and } \sum_{i=1}^{15} [n_i] \leq 1 \tag{4.9}$$

This system of Equations 4.8 and 4.9 can be solved to calculate all of the n_i . Therefore, by solving for the fifteen n_i at all fifteen voltages for each sign of charge, a drop charge distribution with thirty charge bins for both signs of charge will be obtained at each drop size. The final results are drop charge distributions for a total of fifteen drop sizes.

The system is constrained, so it cannot be solved algebraically. Instead, a genetic algorithm [67] was used to find optimal solutions to the problem. The genetic algorithm started with a population of 1,000 random-guess charge distributions, and calculated $F_{theoretical}$ for each guess. The guesses where $F_{theoretical}$ was closest to $F_{measured}$ were selected from the first population and used to create the next generation of 1,000

distributions. The algorithm continued this repeating process for 500 generations, and the best-fitting charge distribution was selected from the 500th generation as the final result. However, genetic algorithms are non-deterministic: they do not necessarily give the same final result every time the model is run. Additionally, the measured values of $F_{measured}$ are the means of multiple measurements and contain uncertainty as shown in Figure 4.4. To account for these two sources of uncertainty (data inversion variability and input data uncertainty), we performed a 100 sample Monte Carlo analysis where values of $F_{measured}$ were pulled from a random normal distribution centered at the mean of the measured values of $F_{measured}$ with a standard deviation matching the standard deviation of the $F_{measured}$ measurements. Each of these randomly selected sets of values of $F_{measured}$ was processed through the genetic algorithm above to give an estimate of the uncertainty in the calculated charge distributions at each droplet size. This uncertainty in the model output reflects both the uncertainty in the input measurements and the uncertainty in the genetic algorithm solution, so it is referred to as the combined measurement and model uncertainty.

In order to estimate the accuracy of the data inversion process using genetic algorithm, a known charge distribution for a specific drop size was used to challenge the model. The percent error (*%Error*) was calculated to compare the results from data inversion based on the average of 100 repeats ($\overline{ModelPrediction}$) and the known charge distribution (*Actual*) at each charge level, as in Equation 4.10:

$$\%Error = \frac{|\overline{ModelPrediction} - Actual|}{Actual} \times 100 \quad (4.10)$$

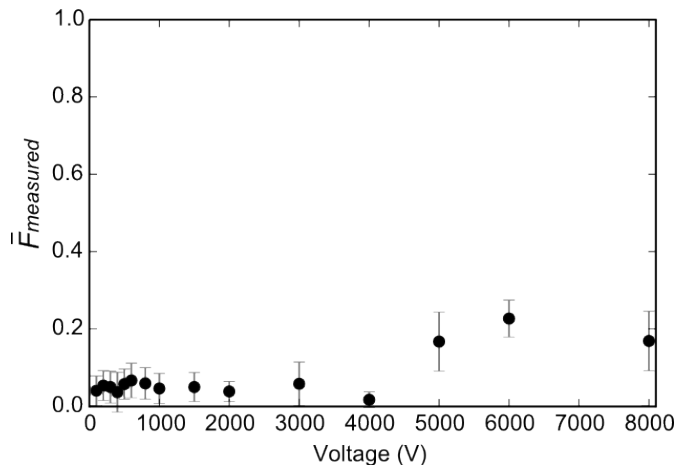


Figure 4.4: Measured Drop Penetration Fractions ($F_{measured}$, mean \pm sd) at fifteen positive voltage levels for 80 μ m plain water spray drops using 10 cm electrical plates.

4.3.5 Experiment and Statistic Design

In this study, seven surfactant solutions, including 10^{-4} M and 10^{-6} M of anionic surfactant, 10^{-4} M and 10^{-6} M of nonionic surfactant, 10^{-4} M and 10^{-6} M of cationic surfactant solutions and plain water were tested for both 10 and 40 cm electrode lengths. Fifteen voltages for both negative and positive signs were applied for both electrode lengths. A total of 540 tests were conducted. Five repeats were conducted within each test. After the data inversion described above was performed to calculate drop charge distributions, descriptive statistics, analysis of variance (ANOVA), and multiple comparison (Scheffé test and Tukey HDS test) were used to analyze the effects of type of surfactant on averaged drop size and drop charge level. Regression and analysis of covariance (ANCOVA) was used to analyze the effects of drop size on drop charge level. The statistical analysis was performed by R project version 2.13.0 [56].

4.3.6 Uncertainty

Coefficients of variation of the mean fractions of charged spray drops (n_i) based on 100 model repetitions were used to compare the combined measurement and model uncertainty between the measurements of 10 cm and 40 cm electrical plates at different drop sizes. However, coefficients of variation were not good parameters to represent uncertainty here because in many measurements the n_i were mostly zeros or close to zero in order to fully cover a wide drop charge range, and thus the coefficients of variation could be very large and did not reflect the real magnitude of uncertainty. Therefore, the coefficients of variation in this study were only used to compare the combined measurement and model uncertainty between the two electrical plates among the n_i that mostly contained non-zero measurements, i.e. the coefficient of variations were compared among the fractions of charged drops that were greater than 10%. The uncertainty of model it self was calculated as

4.4 Results

4.4.1 Spray Drop Size Distribution

The overall drop size distributions for seven test spray solutions as measured at the top of settling chamber are shown in Figure 4.5. The arithmetic average spray drop sizes ranged from 114.2 to 137.0 μm . The geometric mean and standard deviation of tested spray drop sizes are shown in Table 4.2.

ANOVA tests indicated that the average drop sizes among all seven tested sprays were not the same ($p < 0.0001$). According the Scheffé test, plain water had the smallest average drop size among all tested spray drops. Cationic and nonionic surfactant-containing sprays had smaller drop sizes than anionic surfactant-containing sprays,

Table 4.2: The Geometric Mean and Standard Deviation of Tested Spray Drop Sizes

Spray Type	Geometric Mean	Geometric SD
High conc. Nonionic	103.3 ^C	1.95
Low conc. Nonionic	101.2 ^D	1.93
High conc. Cationic	101.3 ^D	1.90
Low conc. Cationic	95.6 ^E	1.93
High conc. Anionic	111.1 ^A	1.98
Low conc. Anionic	106.0 ^B	1.97
Water	91.7 ^F	1.95

Means with different letters are significantly different (Scheffé test, $p < 0.05$).

but the concentration also significantly impacted the drop size. Anionic surfactant-containing sprays had the largest average drop sizes among all tested surfactants, and the average drop size of high concentration anionic surfactant-containing spray was significantly larger than low concentration anionic surfactant-containing spray.

4.4.2 Spray Drop Charge Distribution

Impact of surfactant solution type on charge distribution

The drop charge distributions for the seven test spray solutions, measured using both 10 cm and 40 cm electrical plates, are shown in Figures 4.6 and 4.7 respectively. Ten centimeter electrical plates were able to measure the full charge range for drops up to about 100 μm in diameter, with a charge range from 1 to about 68,000 charges per drop, while 40 cm electrical plates were able to measure the full charge range for the larger drops, with a maximum charge up to about 960,000 charges per drop. The spray drop charge level was significantly impacted by surfactant solution type ($p < 0.0001$).

Two factors are of interest in the drop charge distributions shown in Figures 4.6 and 4.7: the magnitude of charges per drop for both positive and negative charge as

shown by the width of shading in the vertical direction, and the fractions of drops with positive and negative charge as shown as the darkness of the shading. Figure 4.6 shows that both high and low concentration nonionic and cationic surfactant-containing sprays, as well as plain water spray, tended to have a larger fraction of drops with positive charge than with negative charge, indicating an average net positive charge. However, the differences for cationic surfactant-containing sprays were not as large as for nonionic surfactant-containing sprays and water spray. Both high and low concentration anionic surfactant-containing sprays, especially high concentration anionic surfactant-containing spray, tend to have more drops with negative charge than with positive charge, indicating an average net negative charge. In addition, high and low concentration nonionic and anionic surfactant-containing sprays had similar magnitude of charges per drop for both positively- and negatively- charged drops at each drop size, but cationic surfactant-containing sprays and water spray drops tended to carry more positive charges per drop than negative charges at most drop sizes.

Figure 4.7 shows similar results for the fractions of charged drops, except that high and low concentration cationic surfactant-containing sprays tended to have a larger fraction of drops with negative charge than with positive charge, but a larger magnitude of positive charges per drop than of negative charges per drop. Low concentration anionic surfactant-containing sprays tended to have smaller magnitude of charges per drop in both signs of charge than the other sprays. In addition, all the spray solutions tended to carry a larger magnitude of positive charges per drop than of negative charges per drop at many drop sizes. Compared to the data measured using the 10 cm electrical plates, the 40 cm electrical plates produced smaller fractions of charged drops at each drop size but the range of measured drop charges was larger.

The total fractions of drops with positive and negative charge across all fifteen charge levels were separately compared among the seven tested sprays in Table 4.3. For the results obtained with 10 cm plates, the total fractions of positively charged drops were similar among all sprays ($p=0.235$), but the total fractions of negatively charged drop were not all the same ($p=0.038$). However, Tukey's HSD test for multiple comparisons of total fractions of positively- and negatively-charged drops did not find significant difference when comparing individual sprays because of its conservative nature that attempts to control the overall alpha level.

The total fractions of drops with either positive or negative charges were not all the same among tested surfactant types ($p<0.001$) when using the results obtained with 40 cm electrical plates, as shown in Table 4.3. Tukey's HSD test suggests that cationic and anionic surfactant-containing sprays with both high and low concentration had significantly more negatively charged drops than nonionic surfactant-containing sprays with both high and low concentration. High concentration cationic and low concentration anionic surfactant-containing sprays had significantly more negatively charged drops than water spray.

The total fractions of positively and negatively charged drops were also compared for each spray type in Table 4.3 as p-values in each row, as shown in 4th and 7th columns. The charge distribution measured by 10 cm electrical plates indicated that the total fractions of positively charged drops for high and low concentration nonionic surfactant-containing sprays and plain water spray were significantly higher than the total fractions of negatively charged drops, with about as twice as many positively than negatively charged drops. The total fractions of positively charged drops for low and high concentration cationic surfactant-containing sprays were also higher than the total fractions of negatively charged drops, but these differences were not significant. Low and high concentration anionic surfactant-containing sprays had higher

total fractions of negatively charged drops than positively charged drops, but the differences were also not significant. The results given by 40 cm electrical plates were similar, except that the total fractions of negatively charged drops for low and high concentration cationic surfactant-containing sprays were significantly higher than the total fractions of positively charged drops.

This observation suggests that nonionic surfactant-containing sprays and water spray tend to have significantly more positively charged drops, while anionic surfactant-containing spray may contain more negatively charged drops. There were inconsistencies regarding whether cationic surfactant-containing spray contains more positively or negatively charged drops, but it is likely that the cationic spray had more negatively charged drops because the results given by 40 cm electrical plates, which showed more negative charges, were statistically significant, whereas the results given by the 10 cm electrical plates were not statistically significant.

Impact of drop size on charge distribution

Drop charge level was associated with drop size among all spray types. Drop charge level was observed to be significantly higher among larger size drops ($p < 0.0001$). Figure 4.8 shows average drop charge for both positively and negatively charged drops as a function of drop size. Nonionic surfactant-containing sprays had higher positive charges on smaller drops, and higher negative charges on larger drops. However, cationic and anionic surfactant-containing sprays and plain water spray had higher positive charges on most sizes of drops

Uncertainty of the drop charge distribution measurement

The coefficients of variation for measurements using 10 cm electrical plates were evaluated for 20-130 μm drops, as shown in Figure 4.9. The coefficients of variation decreased when the drop size increased from 20 to 40 μm , but increased when the drop size increased from 40 to 130 μm . The average coefficient of variation for drops ≤ 90 μm was 63.9%, and the average coefficient of variation for drops > 90 μm was 73.4%, indicating 10 cm electrical plates were most suitable for measuring charge distribution for smaller drops. The average coefficient of variation among high and low concentration anionic and low concentration nonionic surfactant-containing sprays and water spray, especially the latter, were smaller than among high and low concentration cationic and high concentration nonionic surfactant-containing sprays (\overline{CV} =62.0%, 63.4%, 61.6%, and 60.4%, respectively, as compared to \overline{CV} =68.4%, 67.0%, and 72.2%, respectively).

The coefficients of variation for measurements using 40 cm electrical plates were evaluated for 130-320 μm drops, as shown in Figure 4.10. The coefficients of variation increased as the drop size increased. The average coefficient of variation for drops between 130 to 270 μm was 68.0%, but the average coefficient of variation for drops >270 μm in diameter was 84.1%, indicating 40 cm electrical plates were more suitable for measuring charge distribution for large drops with a diameter between 130 to 270 μm , but may be less suitable for measuring charge for drops larger than 270 μm in diameter. The coefficients of variation among both low and high concentration anionic and high and low concentration cationic surfactant-containing sprays were smaller than among high and low concentration nonionic surfactant-containing sprays and water spray (\overline{CV} =61.2%, 55.5%, 50.8%, and 60.8%, respectively, as compared to \overline{CV} =66.2%, 71.4%, and 68.5%, respectively).

The average coefficients of variation for measurements using 10 cm electrical plates

and using 40 cm electrical plates similar (\overline{CV} =64.6% and 63.2%, respectively). However, the difference in drop size had a smaller impact on coefficients of variation for measurements using 10 cm electrical plates than for those using 40 cm electrical plates across all drop charge levels.

4.4.3 Net Average Charge

Table 4.4 shows net average charge per drop for each spray type. The net charge measured by 10 cm electrical plates indicated that both low and high concentration nonionic and cationic surfactant-containing sprays, as well as plain water spray, carried net positive charges on average, whereas both low and high concentration anionic surfactant-containing sprays carried net negative charges on average. The net charge measured by 40 cm electrical plates indicated that only high concentration anionic surfactant-containing spray carried a net negative charge, while all other sprays carried net positive charges. In addition, both low and high concentration nonionic surfactant-containing sprays, as well as plain water spray, had higher positive charge levels than low and high concentration cationic surfactant-containing sprays. For both nonionic and cationic surfactant-containing sprays, low concentration sprays had higher positive charge levels than high concentration sprays. High concentration anionic surfactant-containing spray had higher negative charge than low concentration anionic surfactant-containing spray.

4.5 Discussion

4.5.1 Drop Size Distribution

The average spray drop Sauter mean diameter was approximately 190 μm across tested spray solutions, which is consistent but towards the large end of the observation of 90 to 210 μm by Pollock and Organiscak [18]. Although this research adopted the same nozzle type, cone angle, water flow and pressure as that in Pollock and Organiscak's study, three nozzles rather than a single nozzle were used in this study to maximize the spray area. The spray plumes of the three nozzles overlapped and therefore may have caused more collisions and coalescence. This could result in an increased Sauter mean diameter [68].

The arithmetic average spray drop diameter ranged from 114.2 to 137.0 μm for the tested sprays. However, this size range may have been overestimated for several reasons. First, the location of the measurement influences the measured size distribution. Drops tend to be smallest around the center of the plume close to the nozzle orifice [69]. Our measurements, however, took place at the center of the plume about 50-60 cm from nozzle opening. Second, small drops tend to follow the air flow in a wind tunnel, rather than fall into the measurement area. Third, small drops evaporate more quickly than large drops, so fewer of them would be measured as they traveled into the measurement area. Finally, the laser drop sizer used in this research had a minimum size limit of 20 μm . However, this limitation should not significantly impact the size distributions reported here because according to the drop size distribution shown in Figure 4.5 it is likely that only a small fraction of spray drops were smaller than 20 μm .

There were statistically significant differences in drop sizes among the seven tested sprays ($p < 0.0001$). Water spray drops had a smaller average drop size (114.2 μm)

than did surfactant-containing spray drops (118.3-137.0 μm). Cationic surfactant-containing spray had the smallest average drop size (121.0 μm) among surfactant-containing sprays, while anionic surfactant-containing spray had the largest average drop size (134.3 μm) ($p < 0.0001$). Based on these results, adding surfactant into spray water did not decrease the average drop size.

4.5.2 Drop Charge Distribution

Fraction of drop with positive and negative charges

The fraction of drops with positive and negative charges can vary among different surfactant-containing sprays. Tukey's HSD tests (Table 4.3) for multiple comparison of the total fractions of drops with positive or negative charge indicate that major differences were seen among nonionic and cationic surfactant-containing sprays and water spray, but not for anionic surfactant-containing sprays. In addition, these differences also tend to be observed when using 40 cm electrical plates. This result suggests that the ionic surfactant sprays had significant impacts on the ability of spray drops to carry negative charges, and this impact may be more significant among larger drops with high charge levels measured by the 40 cm electrical plates. Nonionic surfactant spray with the concentrations tested here may significantly impact the ability of spray drops to carry positive charges. Compared to 40 cm electrical plates, 10 cm electrical plates did not give a full range of charge measurements for larger drops. Therefore, the missing data for highly charged large drops may affect the results reported by the 10 cm electrical plates.

The total fractions of positively and negatively charged drops for each spray type in Table 4.3 also suggests that nonionic surfactant-containing sprays and water spray contained significantly more positively charged drops than negatively charged drops.

Although not statistically significant, anionic surfactant-containing sprays had more negatively charged drops than positively charged drops. These results are consistent with the surfactants used in Polat *et al.* [28]. However, these prior results differ from ours in the ratios of positively and negatively charged drops among the different sprays. This difference may be due to the limited number of individual drops measured by Polat *et al.*, and may also be due to experimental error, which will be discussed below. Although there was inconsistency in the sign of cationic surfactant-containing spray drop charge in our tests, our results may not support the observation by Polat *et al.* that they contained more positively charged drops. Our findings suggest that the surfactant classification, such as nonionic, cationic and anionic, may not be the only determinant for the sign of drop charge. Other factors, such as drop size or the spray process, may also impact the spray drop charge.

Drop charge level

Drop charge level was positively associated with drop size ($p < 0.0001$). Overall, there was an apparent power-law relationship between average drop charge and drop size for drops smaller than 140 μm for all spray solutions, as shown in Figure 4.8. The power-law relationship is less apparent for drops larger than 140 μm , because the measurements by 10 cm electrical plates did not fully cover the drop charge range for drops larger than 80 μm in diameter, the drop charge levels were likely underestimated with these plates for drops larger than 80 μm .

The average drop charges for positively- and negatively- charged drops differ among different spray solutions (Table 4.3). Both high and low concentration cationic surfactant-containing sprays had higher average drop charges for positively- charged drops among most drop sizes. High and low concentration anionic surfactant-containing sprays and water spray also had higher average drop charges on positively-charged drops

than on negatively-charged drops. High and low concentration nonionic surfactant-containing sprays, however, tended to have higher average drop charge on negatively-charged drops than on positively-charged drops. This observation was not consistent with the net drop charge levels among spray solutions (Table 4.4), suggesting that the differences in net drop charge levels may be mainly caused by differences in the fractions of positively- and negatively-charged drops among spray solutions rather than the magnitude of positive and negative charges per drop.

Net average charge

The net drop charge levels averaged across all drop sizes were different among the tested sprays. The measured charge fractions may explain the observed differences among the sprays. Although cationic surfactant-containing sprays had significantly more negatively-charged drops measured by 40 cm plates (Table 4.3), the net drop charges of cationic surfactant-containing sprays measured using the 40 cm plates (Table 4.4) were positive. This observation suggests that, in addition to the fractions of positively- and negatively-charged drops, drop charge level also played an important role on affecting the drop net charge.

4.5.3 Limitations

The spray drop charge measurement system developed in this study allows the measurement of spray drop electrostatic charge. However, there are several limitations that may impact the accuracy and precision of the measurements.

The design of the spray drop charge measurement system was based on the assumption that within the electrical field all the horizontal movements of drops are caused by the electrical force. Although the sealed testing chamber was designed for limiting air movement, and a narrow metal tube was used to prevent the drops with

initial horizontal movements from falling into the drop classifier, drops could still experience horizontal movements caused by the movement of air in the wind tunnel and settling chamber. Those horizontal movements of drops could cause an underestimate of the Drop Penetration Fraction. Because the Drop Penetration Fraction depends on both horizontal movement and plate length, this effect could impact the results from the 40 cm electrical plates more than from 10 cm electrical plates. This effect may explain why the drop charge distributions measured by 10 cm and 40 cm electrical plates are not the same for the same drop sizes.

Owing to limited space for the experimental setup, the spray drop charge measurement system was designed to only measure drops smaller than 300 μm in diameter. Additionally, the spray drop charge measurement system was designed to measure a range of sizes and charges based on previously reported charge distributions [18, 28]. However, the measurements in this study found a larger range of drop sizes and charges. Therefore, the 10 cm electrical plates were less accurate to measure drops $>90 \mu\text{m}$ in diameter, and 40 cm electrical plates were less accurate to measure drops $>270 \mu\text{m}$ in diameter.

In addition, the data inversion process using the genetic algorithm also causes modeling error. An example of a comparison between a known charge distribution and the model prediction of that charge distribution is shown in Figure 4.11. The percent error between the model prediction and the actual charge distribution was approximately 20% across all charge levels, which is believed to be sufficiently accurate for the purposes of this study.

Regardless of these limitations, the spray drop charge measurement system can provide important information of drop charge distribution. Information found here can be used to improve future spray drop charge measurement system designs.

There are steps that future studies could take to reduce the inaccuracy caused by

the horizontal movements of drops. First, either reducing the nozzle pressure or the wind tunnel air velocity could decrease the horizontal movements of drops in the settling chamber by lowering the average horizontal velocity of the drops in the wind tunnel. However, reducing the nozzle pressure may also alter the drop size and charge. Second, a correction factor could be calculated to account for the drops lost due to horizontal movements. This correction factor could be calculated by comparing the total number of drops at the bottom of the drop classifier (i.e., across the entire width of $d = 14$ mm) when there is no electrical field created to the total number of drops at the bottom of the drop inlet (i.e., across the entire width of $d_1 = 6$ mm).

To improve the inaccuracy of measurement on larger and higher charged drops, using a longer settling chamber could allow drops larger than $300\ \mu\text{m}$ to reach their terminal settling velocity. In addition, using longer electrical plates could cover the full drop charge range for drops $> 270\ \mu\text{m}$ in diameter.

4.6 Conclusion

Low and high concentration nonionic and cationic surfactant-containing sprays and water spray carry net positive charge on average. However, increasing the concentration of cationic surfactant-containing sprays from 10^{-6} M to 10^{-4} M did not increase net positive charge in spray drops. High concentration anionic surfactant-containing spray carries net negative charge on average.

The spray drop charge measurement system described here can be used to measure spray drop electrostatic charge for drops smaller than $300\ \mu\text{m}$ in diameter. The strengths of this drop charge measurement system are that it allows the measurement of the drop charge and size distribution for many drops at once, and that it allows

the measurement of a relatively wide drop size and charge range. The major limitations are that the horizontal movements of drops that are not caused by the electrical force in the drop classifier cause inaccuracies in measurements, and that the charge distribution of drops greater than 300 μm in diameter are not accurate at the current settings. Future design improvements may improve accuracy by conducting additional measurements to more accurately measure the Drop Penetration Fraction and by decreasing the air velocity in the test system.

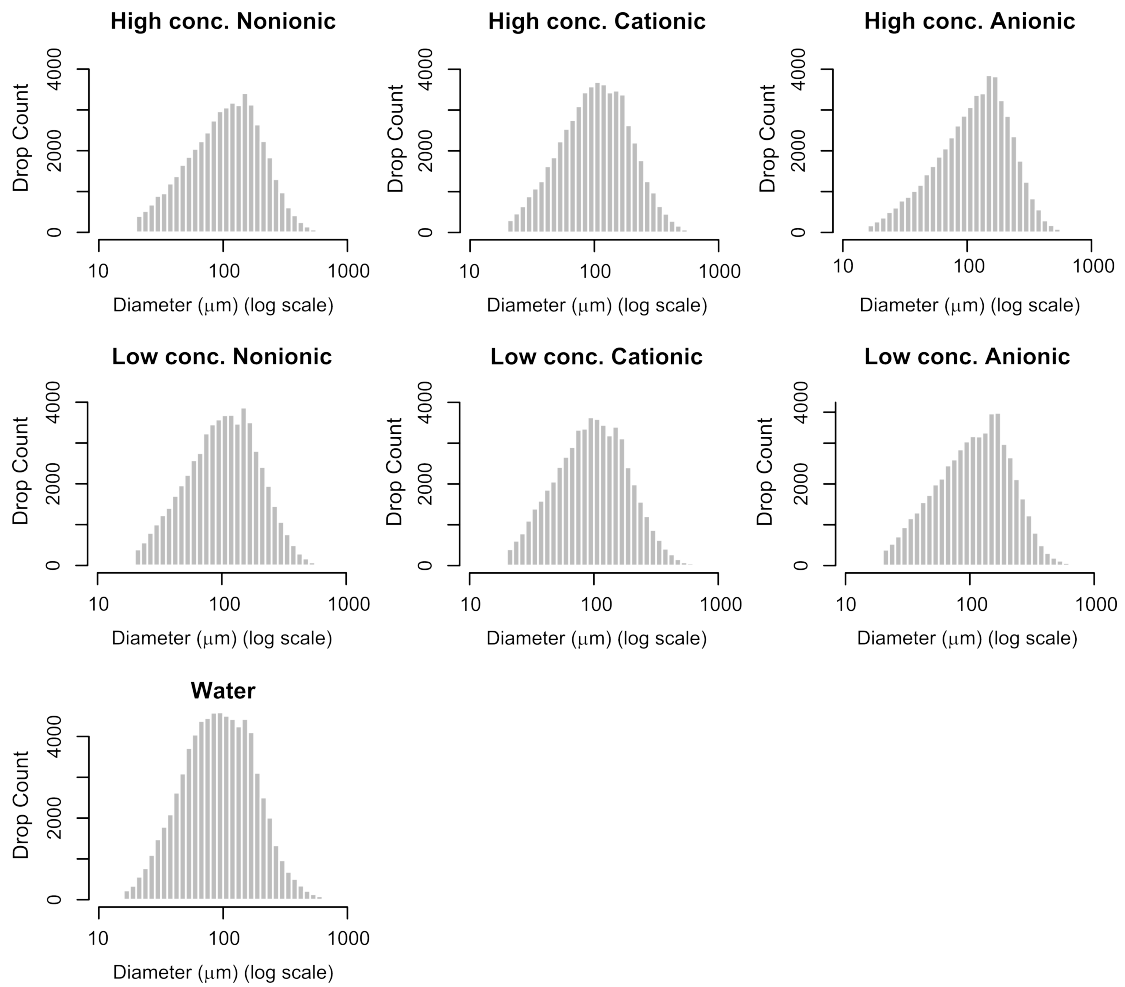


Figure 4.5: Histograms of drop size distributions by drop count for seven tested spray solutions

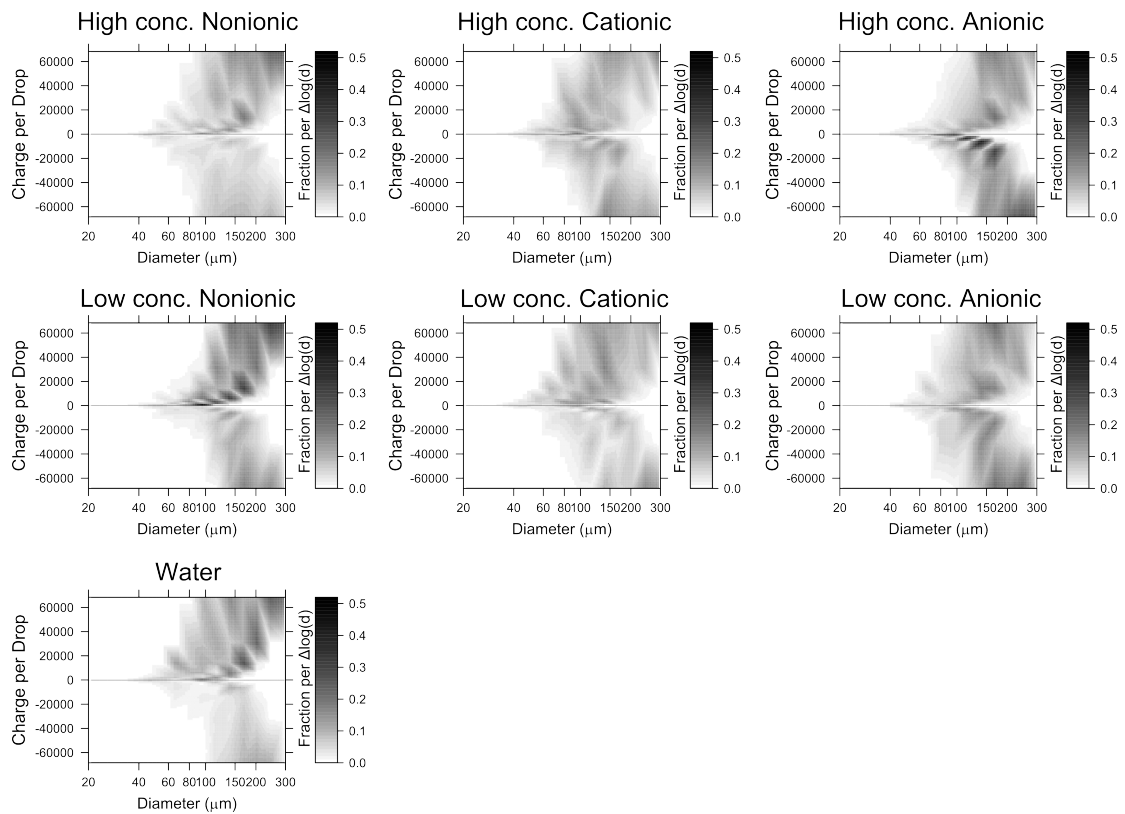


Figure 4.6: Mean fractions of charged spray drops at different drop sizes and charge levels measured by 10 cm electrical plates, normalized by $\Delta\log(d)$

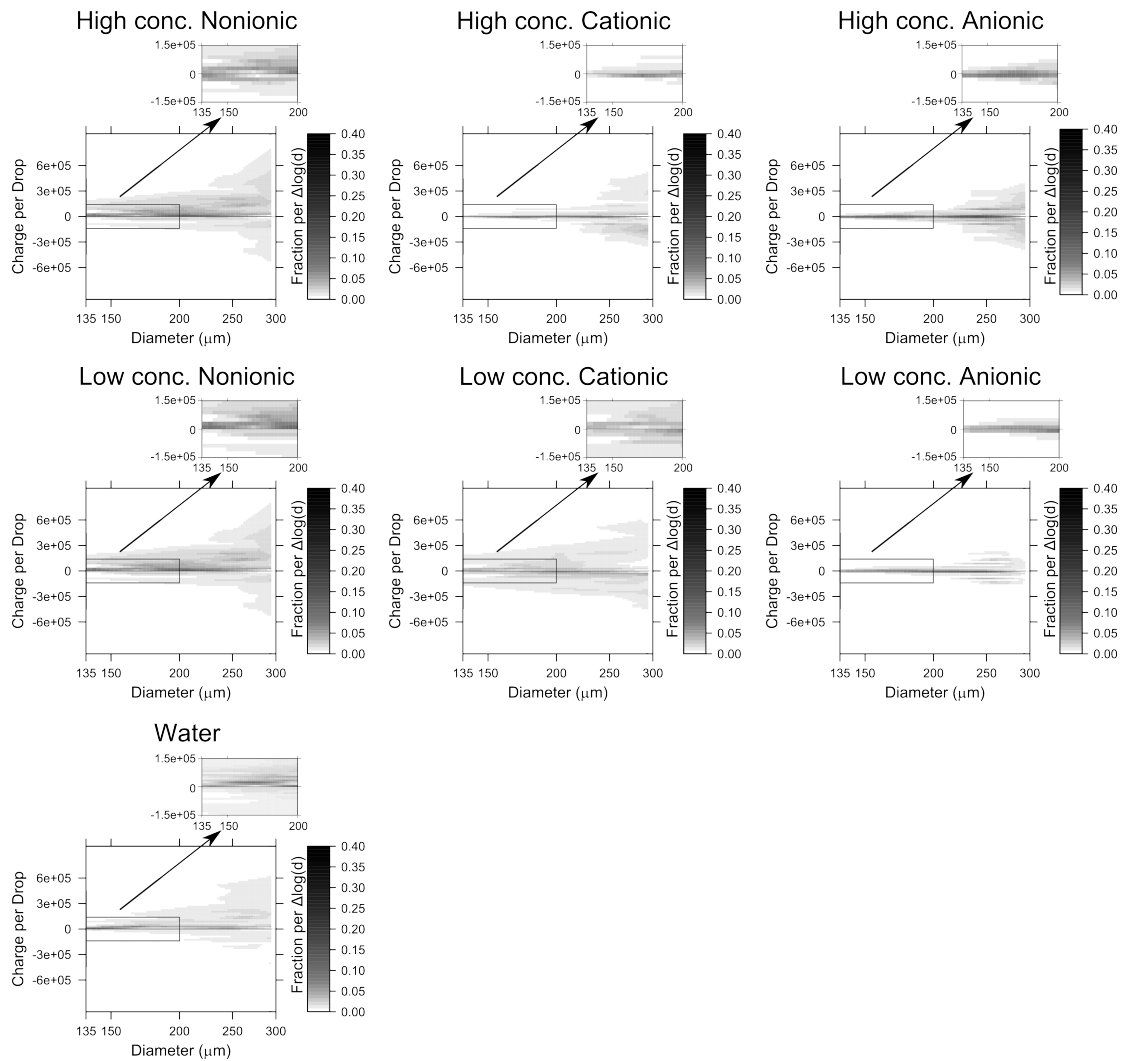


Figure 4.7: Mean fractions of charged spray drops at different drop sizes and charge levels measured by 40 cm electrical plates, normalized by $\Delta \log(d)$

Table 4.3: Comparison of the Total Fractions of Positively- and Negatively-Charged Drops across All Fifteen Charge Levels of Different Spray Solutions for Both 10 cm and 40 cm Electrical Plates

Spray solutions	10 cm electrical plates			40 cm electrical plates		
	Total fraction of positively charged drops	Total fraction of negatively charged drops	p-value (*p<0.05)	Total fraction of positively charged drops	Total fraction of negatively charged drops	p-value (*p<0.05)
High conc. Nonionic	0.57 ^A	0.32 ^A	0.003*	0.42 ^B	0.29 ^C	0.010*
Low conc. Nonionic	0.62 ^A	0.33 ^A	0.002*	0.62 ^A	0.21 ^C	<0.0001*
High conc. Cationic	0.48 ^A	0.41 ^A	0.303	0.38 ^B	0.57 ^A	0.001*
Low conc. Cationic	0.51 ^A	0.38 ^A	0.055	0.36 ^B	0.45 ^{AB}	0.043*
High conc. Anionic	0.45 ^A	0.48 ^A	0.776	0.42 ^B	0.51 ^{AB}	0.146
Low conc. Anionic	0.44 ^A	0.47 ^A	0.675	0.42 ^B	0.53 ^A	0.111
Water	0.63 ^A	0.30 ^A	0.001*	0.50 ^{AB}	0.35 ^{BC}	0.005*

In columns 2, 3, 5, and 6, means with different letters are significantly different (Tukey's HSD test, p<0.05). * indicates the total fractions of positively charged drops and the total fractions of negatively charged drops are significantly different (t-test, p<0.05).

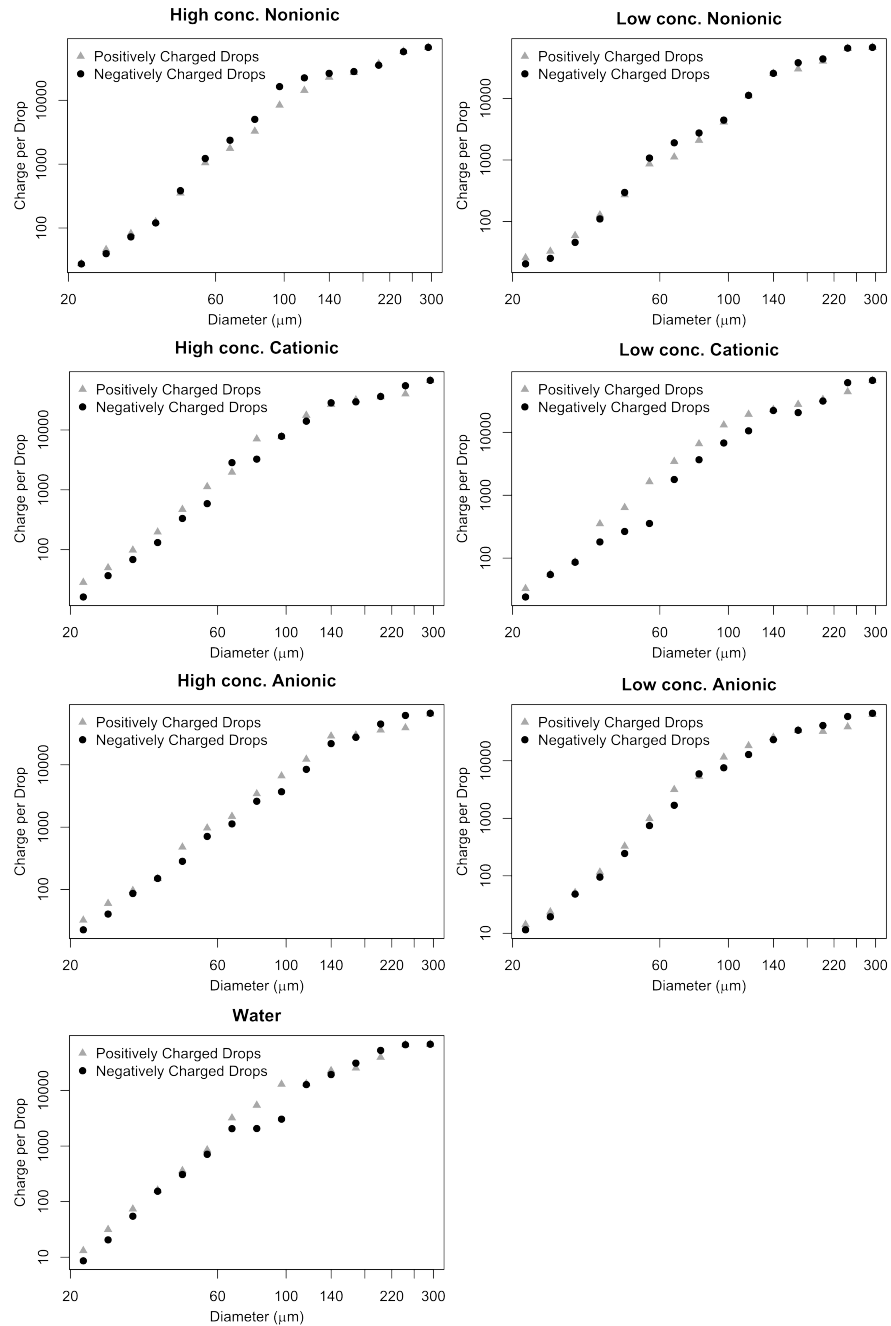


Figure 4.8: The average drop charge as a function of drop diameter for both positively- and negatively- charged drops, measured by 10 cm electrical plates

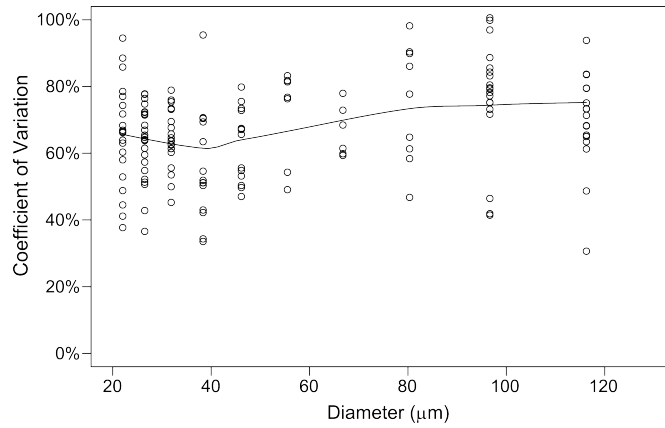


Figure 4.9: Coefficients of variation vs. drop diameter across all drop charge levels and spray types with a LOESS regression line, measured by 10 cm electrical plates

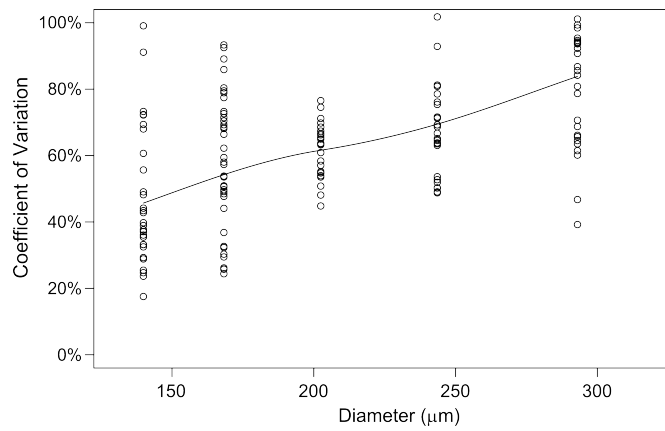


Figure 4.10: Coefficients of variation vs. drop diameter across all drop charge levels and spray types with a LOESS regression line, measured by 40 cm electrical plates

Table 4.4: Net Average Charge per Drop for Seven Tested Sprays Measured by Both 10 and 40 cm Electrical Plates

Measurement	Water	Low conc. Non-ionic	High conc. Non-ionic	Low conc. Cationic	High conc. Cationic	Low conc. An-ionic	High conc. An-ionic
10 cm electrical plates	5241	4912	3645	2957	937	-245	-1478
40 cm electrical plates	3343	5757	3692	790	25	194	-77

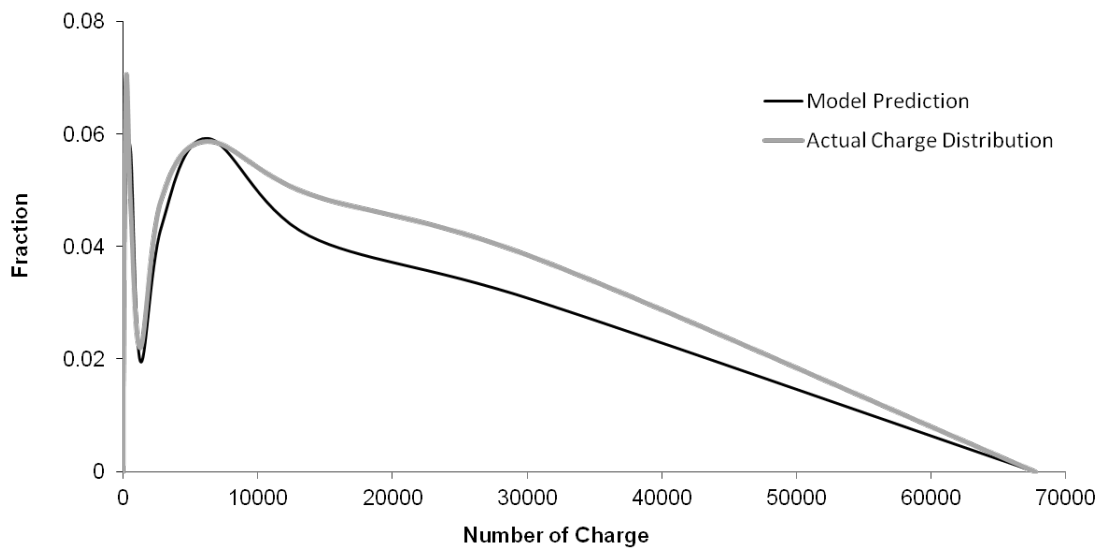


Figure 4.11: An example of a comparison between a charge distribution and its corresponding model prediction

Chapter 5

Statistical Analysis for Factors that Influence the Surfactant-Containing Spray Control of Respirable Particles

5.1 Summary

Adding surfactant into spray water can cause changes in spray solution surface tension, drop size and drop charge level. These changes can all affect spray collection efficiency. This study collected data on spray solution surface tension, and analyzed spray solution surface tension and previously collected data on spray collection efficiency, particle size, particle charge, drop size, and drop charge to determine how changes in drop charge levels caused by adding surfactant can cause changes in spray collection efficiency after controlling for other drop parameters. The results suggested a positive correlation between spray collection efficiency and the drop charge magnitude after controlling for spray solution surface tension and drop size. The interaction

of the particle charge magnitude and drop charge magnitude may also affect spray collection. The increase in the particle charge magnitude may even enhance the impact of the drop charge magnitude on collection efficiency among weakly-charged particles.

5.2 Introduction

Chapters 2 and 3 discussed the effects of surfactant concentration, surfactant classification, and dust particle charge level on collection efficiency of respirable PSL and coal dust particles by surfactant-containing spray. However, as discussed in Chapter 1, respirable particle capture efficiency could also be affected by spray surface tension, drop size, and drop charge.

Previous studies have indicated that the presence of surfactants in spray water could change the spray drop size and collection efficiency [16]. Bughdadi reported a reduction in scrubber spray drop sizes with the presence of 0.1% nonionic surfactant Triton CF-10 (cited in [19]). Woffinden *et al.* [19] used a scrubber model to predict that the use of a surfactant would decrease drop diameter in particle scrubbers by 30%, and hypothesized that smaller drop diameters would lead to higher collection efficiency. Ellis *et al.* [20] suggested that adding nonionic surfactants (0.5% Fatty alcohol ethoxylates, Polyoxyethylene sorbitan fatty acid ester, Alkoxylated ethylenediamine, or Alkyl polyglycoside) could reduce the volume median diameter of spray drops by up to 17%. However, Tien and Kim [11] did not observe significant drop size changes with the addition of nonionic surfactant (0.5% Surfynol440 and Plurafac RA 43).

Many studies have attributed the improvement of surfactant-containing spray or scrubber collection efficiency to the reduction of spray solution surface tension because lower surface tension in the drop leads to more effective wetting and allows

drops to contain more particles. McCully *et al.* [21] reported that less wettable airborne particles were less likely to be removed by raindrops. Bughdadi reported that adding 0.1% nonionic surfactant Triton CF-10 improved the overall particle collection efficiency by increasing the penetration of particles into scrubber droplets (cited in [19]). Chander *et al.* [27] reported that nonionic surfactants (both $>3 \times 10^{-4}$ M Triton X-100 and $>1 \times 10^{-4}$ M Triton N-101) could be used as effective wetting agents for coal dust. Chander *et al.* [22, 12] also pointed out that decreased surface tension could improve drop collection effectiveness especially in high dust concentration situations. Li *et al.* [26] found that anionic surfactant (0.2% sodium dodecyl sulfate) enhanced wetting performance significantly on respirable coal mine dust. Tien and Kim [11] also concluded that nonionic surfactants may have better wetting qualities on coal dust because they have a balanced structure consisting of hydrophilic head groups and hydrophobic tail groups. Wetting performance can be improved by up to 215% using sodium hydroxide and potassium hydroxide surfactants [11].

The charge of drops in a spray or scrubber can also affect collection efficiency. Kraemer and Johnstone [70] showed that the collision efficiency of small dust particles with charged collectors depended on Coulombic interaction. Increased water drop charge increases particle removal efficiency, especially for oppositely-charged particles [38].

Adding surfactant to spray systems could significantly alter the charge on the drops, and the sign and the magnitude of the dominant drop charge could depend on the sign of the charge group in surfactant's hydrophilic head [28]. In general, adding anionic and cationic surfactants can cause spray drops to be more negatively and positively charged, respectively. The charge level on drops can reach its peak when the concentration of ionic surfactant is approximately 10-100 times more diluted than the critical micelle concentration (CMC) [30]. Nonionic surfactants, such as Triton X-100,

cause more positive charges on spray drops, and the charge level on these drops increases as the surfactant concentration increases [28]. Because the drop charge level impacts the spray collection efficiency, it is important to have quantitative information on the impact of the types and concentrations of surfactants used in this study on drop charge rather than qualitative estimate based on the surfactant classification.

The objective of this study is to investigate the impact of changes in spray drop electrical charge caused by adding surfactant on dust capture efficiency. However, the presence of surfactant in spray water could affect the spray collection efficiency by multiple mechanisms. Therefore, it is important to control for other factors that may be changed by surfactant addition, such as spray drop surface tension and drop size, in order to understand the relationship between spray drop electrostatic charge and the spray collection of charged respirable dust.

5.3 Methods

To supplement the previously completed measurements of spray collection efficiency and particle parameter, parameters of spray drops from different surfactant solutions were measured including spray drop charge, spray solution surface tension, and drop size. The impact of drop charge on spray collection efficiency was analyzed after statistically controlling for the effects of the other drop parameters.

The methods for measuring overall spray collection efficiency for particles with all charge levels and charge-specific collection efficiency for particles with certain charges have been described in Chapter 2 and 3. Briefly, collection efficiencies for both PSL particles and coal dust were measured by calculating the penetration when different surfactant sprays were applied.

The methods of spray drop size and drop charge measurements have been described in Chapter 4. The sign and magnitude of net average charge were calculated based on the spray drop charge distribution across all drop sizes.

The surface tension of surfactant-containing solutions was measured by the drop-weight method [71]. The surface tension was calculated as

$$\sigma = \sigma_{water} \frac{m}{m_{water}} \quad (5.1)$$

where σ is the surface tension of the surfactant-containing solution, σ_{water} is the surface tension of pure water, m is the total mass of surfactant-containing solution drops, and m_{water} is the total mass of pure water drops. Each weight measurement of pure water and each surfactant-containing solution was the average of three repeats based on 40 drops. The room temperature during the measurements was 22 ± 0.5 °C.

The impact of particle parameters including particle size, aerosol charge condition, and particle charge level, as well as spray drop parameters including spray solution surface tension, spray drop size, and spray drop charge level, on spray collection efficiency were characterized in this study. Aerosol charge condition and particle charge level characterize the charge profile for a group of particles and an individual particle respectively. Therefore, the impact of aerosol charge condition was only associated with overall collection efficiency and the particle charge level was only associated with charge-specific collection efficiency.

According to previous results in Chapters 2 and 3, particle size was the most important factor impacting the spray collection efficiency. Its impact was tested here using simple linear regression. The model for this simple linear regression can be described as

$$\text{Efficiency} = a + b * D_p + E \quad (5.2)$$

where particle diameter (D_p) is the main effect, a is the coefficient for the intercept,

b is the coefficient of for particle diameter, and E is the error term. The impact of each individual factor other than particle size on collection efficiency was tested for significance after controlling the particle size using one-way ANCOVA for categorical variables and multiple linear regression for the continuous variables. The model of multiple linear regression for continuous variables can be described as

$$\text{Efficiency} = a + b_1 * D_p + b_2 * X_2 + E \quad (5.3)$$

where X_2 represents any of the continuous variables being solved for, a is the coefficient for the intercept, b_1 is the coefficient for particle diameter (D_p), b_2 is the coefficient for X_2 , and E is the error term. The one-way ANCOVA model for categorical variables (e.g. spray type) can be described as

$$\text{Efficiency} = a + b * D_p + \sum_{j=1}^{k-1} c_j * Z_j + E \quad (5.4)$$

where a is the coefficient for the intercept, b is the coefficient for particle diameter (D_p), Z is the dummy variable that has various states j which range from 1 to $k - 1$, k is the number of conditions for Z , c_j is the coefficient of for Z_j , and E is the error term. If the impact of the each individual factor was statistically significant, multiple comparison Tukey HSD and Scheffé tests were performed for categorical variables. The Scheffé test was only used for the comparison that had unequal sample size.

Multiple linear regression was used to select a best fit model to determine how the main effects and interaction terms impact the spray collection efficiency. The model of multiple linear regression can be described as

$$\text{Efficiency} = a + \sum b_i * X_i + \sum b_{jk} * X_j * X_k + E \quad (5.5)$$

where a is the coefficient for the intercept, b are the coefficients for continuous variables X , X_i are main effects, $X_j * X_k$ represent interaction terms, and E is the error

term. For overall collection efficiency, independent variables including particle size, aerosol charge condition, spray surface tension, drop size, and drop charge magnitude were initially fitted in the multiple linear regression analysis. For charge-specific collection efficiency, independent variables including particle size, the particle charge magnitude, spray surface tension, drop size, and the drop charge magnitude were initially fitted in the multiple linear regression analysis. Forward selection kept only the independent variables and their interactions that had significant impact on collection efficiency and explained the most collection efficiency-associated variability. Statistical analyses were performed using R project [56].

5.4 Results

5.4.1 Particle Parameters

Particle diameter had the largest impact on overall spray collection efficiency ($p < 0.0001$) for both PSL particles and coal dust across all particle charge levels and surfactant types according to the simple linear regression analysis, as shown in Table 5.1. Overall collection efficiency for PSL and coal dust increased as particle size increased. The overall collection efficiency based on particle number concentration was $21.5\% \pm 9.0\%$ (mean \pm SD) for small respirable size ($0.6 \mu\text{m}$) PSL particles, $58.8\% \pm 12.5\%$ for medium size ($1.0 \mu\text{m}$) particles, and $86.6\% \pm 43.5\%$ for large size ($2.1 \mu\text{m}$) particles across all types of surfactant-containing sprays. Simple linear regression suggested that a $1 \mu\text{m}$ increase in particle size corresponded to an increase in overall collection efficiency of 40% for PSL particle based on the size range from 0.6 to $2.1 \mu\text{m}$ and of about 18% for coal dust based on the size range from 0.58 to $3.2 \mu\text{m}$, as shown in Table 5.1.

However, the overall collection efficiencies for coal dust with various sizes did

Table 5.1: Summary of the Impacts of Individual Factors on Overall Collection Efficiency, Factors in the 2nd to 6th Rows Were Controlled for Particle Size

Factors	Overall Collection Efficiency	
	PSL Particles	Coal Dust
Particle size	Regression coefficient: 0.40 (μm^{-1}) p<0.001	Regression coefficient: 0.18 (μm^{-1}) p<0.001
Aerosol charge condition	p=0.77	p= 0.80
Spray type	p=0.033	p<0.001
Drop size	Regression coefficient: -0.0033 (μm^{-1}) P=0.35	Regression coefficient: -0.0046 (μm^{-1}) p<0.001
Surface tension	Regression coefficient: -0.0041 ((N/m) $^{-1}$) P=0.050	Regression coefficient: -0.0036 ((N/m) $^{-1}$) p<0.001
Drop charge magnitude	Regression coefficient: 0.000010 (# of charge per drop $^{-1}$) P=0.302	Regression coefficient: 0.000020 (# of charge per drop $^{-1}$) p<0.001

not show a simple linear relationship between particle size and overall collection efficiency, as shown in Figure 5.1. The overall collection efficiency increased more rapidly for relatively small particles, diameters from 0.58 to 1.5 μm , and more slowly for particles larger than 1.5 μm in diameter. The variances in the overall collection efficiency were larger among smaller and larger size coal dust particles than medium-sized particles.

Particle size also significantly impacted charge-specific collection efficiency for both PSL particles and coal dust across all particle charge levels and surfactant types according to the simple linear regression analysis (both $p<0.001$), as shown in Table 5.2. This is consistent with theory because electrical effects mainly affect particles smaller than 2 μm , the charge-specific collection efficiencies were analyzed based on the 0.6 and 1.0 μm PSL particles and 0.58 to 1.8 μm coal dust. The regression analysis indicated that a 1 μm increase in particle size would cause an increase in charge-specific collection efficiency of 120% for PSL particle and of about 69% for coal dust

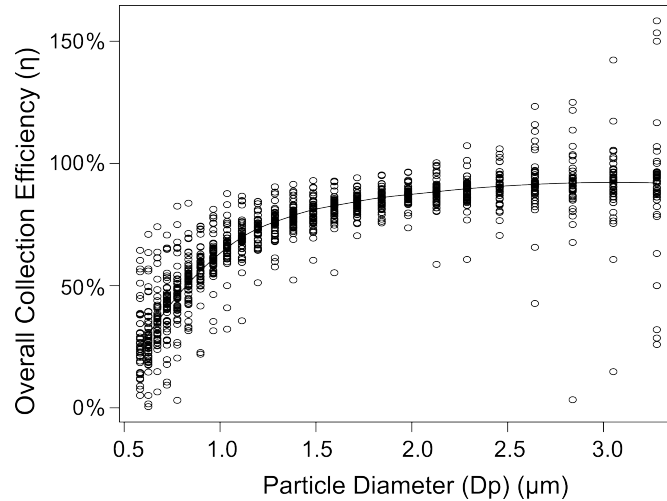


Figure 5.1: Overall collection efficiency (η) vs. coal dust diameter (D_p) across all surfactant sprays and water spray, with a LOESS regression line

(Table 5.2). Because the relationship between particle size and collection efficiency was nonlinear and concave, the regression analysis for PSL particles likely represents an overestimate.

There was no statistical difference in spray collection efficiency among four aerosol charge conditions after controlling for particle size in either PSL particles or coal dust according to an ANCOVA test ($p=0.77$ and $p=0.80$, respectively), as shown in Table 5.1. However, particle charge had a significant effect on charge-specific collection efficiency when the particle size was controlled for in both PSL particles and coal dust (both $p<0.0001$), as shown in Table 5.2. Multiple linear regression suggested that when the particle charge magnitude increased by 1 charge per particle, the charge-specific collection efficiency could increase 0.96% for PSL particles ranging from 4 to 102 charges per particle, and the charge-specific collection efficiency could increase 0.5% for coal dust particles ranging from 3 to 195 charges per particle.

Table 5.2: Summary of the Impacts of Individual Factors on Charge-Specific Collection Efficiency, Factors in the 2nd to 6th Rows Were Controlled for Particle Size

Factors	Charge-Specific Collection Efficiency	
	PSL Particles	Coal Dust
Particle size	Regression coefficient: 1.2 (μm^{-1}) p<0.001	Regression coefficient: 0.69 (μm^{-1}) p<0.001
Particle charge magnitude	Regression coefficient: 0.0096 (# of charge per particle ⁻¹) p<0.001	Regression coefficient: 0.0050 (# of charge per particle ⁻¹) p<0.001
Spray type	p=0.20	p<0.001
Drop size	Regression coefficient: 0.0086 (μm^{-1}) p=0.084	Regression coefficient: 0.0047 (μm^{-1}) p=0.36
Surface tension	Regression coefficient: -0.0093 ((N/m) ⁻¹) p=0.026	Regression coefficient: -0.0095 ((N/m) ⁻¹) p=0.018
Drop charge magnitude	Regression coefficient: -0.000012 (# of charge per drop ⁻¹) p=0.55	Regression coefficient: 0.000059 (# of charge per drop ⁻¹) p=0.016

5.4.2 Drop Parameters

Spray type impacted the effectiveness of overall capture of PSL particles and coal dust when the particle size was controlled for according to an ANCOVA test ($p=0.033$ and $p<0.001$, respectively), as shown in Table 5.1. A multiple comparison using a Tukey HSD test indicated that after controlling for dust size, high concentration nonionic surfactant-containing spray had significantly higher overall collection efficiency than high concentration anionic surfactant-containing spray for PSL particles, and both high concentration nonionic surfactant-containing spray and water spray had significantly higher overall collection efficiency than high concentration anionic surfactant-containing spray for coal dust, as shown in Table 5.3. The overall collection efficiencies among other surfactants or other concentrations did not show significant differences. Spray type also had a significant impact on charge-specific collection efficiency for

coal dust ($p < 0.001$), but not for PSL particles, as shown in Table 5.2. A multiple comparison using a Scheffé test suggested that high concentration nonionic surfactant-containing spray had significantly greater charge-specific collection efficiency than high concentration cationic surfactant-containing spray for coal dust, as shown in Table 5.3.

Table 5.3: Comparison of Mean of Spray Collection Efficiency by Spray Types after Controlling Particle Size

Spray Type	Overall Collection Efficiency (PSL Particles)	Overall Collection Efficiency (Coal Dust)	Charge-Specific Collection Efficiency (Coal Dust)
Water	56.8% ^{ab}	65.6% ^a	53.5% ^{ab}
High conc. Nonionic	64.6% ^a	65.8% ^a	58.0% ^a
High conc. Anionic	43.7% ^b	53.0% ^b	50.4% ^{ab}
High conc. Cationic	49.9% ^{ab}	58.6% ^{ab}	46.6% ^b
Low conc. Nonionic	54.7% ^{ab}	-	-
Low conc. Anionic	60.4% ^{ab}	-	-
Low conc. Cationic	59.6% ^{ab}	-	-

Means of overall collection efficiency with different letters are significantly different by Tukey HSD test ($p < 0.05$), and means of charge-specific collection efficiency with different letters are significantly different by Scheffé test ($p < 0.05$) due to unequal sample size.

Spray type significantly influenced spray solution surface tension, drop size and the drop charge magnitude according to a multivariate regression analysis ($p < 0.0001$). In addition, there were correlations among the impacts of spray solution surface tension, drop size and drop charge. For example, drop size significantly depended on spray solution surface tension ($p = 0.002$), while drop charge significantly depended on spray solution surface tension, drop size, and the interaction between the two (all $p < 0.0001$). Owing to the complication of the interactions among drop parameters, the impact of solution surface tension, drop size and the drop charge magnitude on spray

collection efficiency are reported separately below.

The average drop sizes for different spray types are shown in Table 5.4. The average drop size did not significantly impact the overall collection efficiency for PSL particles after controlling for particle size according to a linear regression analysis ($p=0.35$), as shown in Table 5.1. However, the average drop size significantly impacted the overall collection efficiency for coal dust ($p<0.001$). A linear regression analysis indicated that when the drop size was increased by 1 μm , the overall collection efficiency for coal dust could decrease 0.46% after controlling for particle size (Table 5.1). This result indicates that the overall collection efficiency by water spray may be 11% more than by high concentration anionic surfactant-containing spray owing to a smaller average drop size. Drop size did not significantly impact the charge-specific collection efficiency for both PSL particles and coal dust (Table 5.2).

Table 5.4: Values of Each Drop Parameter for Different Spray Types

Spray Type	Drop Size (μm) (Geometric mean \pm SD)	Surface Tension (N/m)	Net Drop Charge (# of charge per drop)
High conc. Nonionic	103.3 \pm 1.95	47.64	3645
Low conc. Nonionic	101.2 \pm 1.93	68.08	4912
High conc. Cationic	101.3 \pm 1.90	69.75	937
Low conc. Cationic	95.6 \pm 1.93	73.08	2957
High conc. Anionic	111.1 \pm 1.98	73.14	-1478
Low conc. Anionic	106.0 \pm 1.97	72.65	-245
Water	91.7 \pm 1.95	72.30	5241

The average spray surface tensions for different spray types are shown in Table 5.4. Surface tension had a significant impact on overall collection efficiency for both PSL particles and coal dust ($p=0.050$ and $p<0.001$, respectively) after controlling for particle size, as shown in Table 5.1. A linear regression analysis indicated that when the spray surface tension was increased by 1 N/m, the overall collection efficiency for both PSL

particles and coal dust could decrease by approximately 0.4% after controlling the particle size (Table 5.1). Therefore, the overall collection efficiency by high concentration nonionic surfactant-containing spray may be 10% more than by high concentration anionic surfactant-containing spray owing to a lower spray surface tension. Spray surface tension also had a significant impact on charge-specific collection efficiency for PSL particles and coal dust (Table 5.2). When spray surface tension was increased by 1 N/m, the charge-specific efficiency could decrease by approximately 1%, as shown in Table 5.2.

The net drop charges for different spray types are shown in Table 5.4. The magnitude of spray drop charge level, i.e. the absolute value of net drop charge, did not significantly impact overall collection efficiency for PSL particles by controlling particle size ($p=0.302$). However, it significantly impacted the overall collection efficiency for coal dust, so that when the drop charge magnitude was increased by 1 charge per drop, the overall collection efficiency could increase by 0.002%. Therefore, there could be an approximately 11% difference in overall collection efficiency between most highly charged spray drop (water) and most weakly charged spray drop (low concentration anionic) owing to the difference in the magnitude of the spray drop charges. Similar results for charge-specific collection efficiency were seen for coal dust. When the drop charge magnitude was increased by 1 charge per drop, the charge-specific collection efficiency for coal dust could increase by 0.006% (Table 5.2).

5.4.3 Multiple Regression

Impacts of particle size, aerosol charge condition, spray surface tension, drop size, drop charge and their interactions on overall collection efficiency were analyzed by multiple regression. According to model comparison, the best fit model for PSL overall collection efficiency indicated that the collection efficiency was significantly affected

by particle size, drop charge magnitude, and the interaction of surface tension and drop charge magnitude ($p < 0.001$, $p = 0.014$, $p = 0.014$, respectively), as shown in Table 5.5. This model predicted an increase in particle size and the drop charge magnitude could cause an increase in PSL overall collection efficiency after controlling the impact of spray surface tension and drop size. However, the impact of the drop charge magnitude on overall collection efficiency could decrease with increasing spray surface tension. The best fit model can explain about 48% of the variance in PSL overall collection efficiency (Table 5.5). The best fit for coal dust overall collection efficiency indicated that particle size, spray surface tension, and drop size had significant impacts on overall collection efficiency (all $p < 0.001$), as shown in Table 5.6. An increasing particle size and a decreasing spray surface tension or drop size could result in an increase in coal dust overall collection efficiency. However, this model can only explain about 24% of the variance of coal dust overall collection efficiency (Table 5.6).

Table 5.5: Regression Coefficients and Statistical Significance Results for Best Fit Multiple Linear Regression Models of PSL Overall Collection Efficiency

Best fit Model						
Residual Standard Error =0.276, R ² =0.476, Adjusted R ² =0.463, p<0.0001, df=245		Intercept	Main effects and regression coefficients		Interaction terms and regression coefficients	
PSL overall collection efficiency ~ Particle Size+ Drop Size+ Surface Tension+ Drop Charge Magnitude+ Surface Tension: Drop Charge Magnitude + Surface Tension: Drop Size	78.4	Particle Size ^{***}	3.96×10^{-1}	Surface Tension: Drop Size	1.11×10^{-2}	
		Drop Size	-8.45×10^{-1}			
		Surface Tension	-1.02	Surface Tension: Drop Charge Magnitude *	-3.91×10^{-5}	
		Drop Charge Magnitude*	2.70×10^{-3}			

***p<0.001, **0.001≤p<0.01, *0.01≤p<0.05; otherwise, p ≥ 0.05.

Table 5.6: Regression Coefficients and Statistical Significance Results for Best Fit Multiple Linear Regression Models of Coal Dust Overall Collection Efficiency

Best fit Model			
Residual Standard Error =0.328, $R^2=0.237$, Adjusted $R^2=0.235$, $p<0.0001$, $df=1340$	Intercept	Main effects and regression coefficients	Interac- tion terms
Coal dust overall collection efficiency \sim Particle Size+ Drop Size+ Surface Tension	1.28***	Particle Size***	1.83×10^{-1}
		Drop Size***	-5.04×10^{-3}
		Surface Tension***	-3.94×10^{-3}

*** $p<0.001$, ** $0.001 \leq p < 0.01$, * $0.01 \leq p < 0.05$; otherwise, $p \geq 0.05$.

Impacts of particle size, particle charge magnitude, spray surface tension, drop size, drop charge magnitude and their interactions on charge-specific collection efficiency were also analyzed by multiple regression. The best fit model for PSL charge-specific collection efficiency included all the analyzed variables and their interaction terms, as shown in Table 5.7. This model predicted that an increase of particle size, particle charge magnitude and drop charge magnitude could cause an increase in collection efficiency, while an increase in spray surface tension and drop size could cause a decrease in collection efficiency. In addition, the impact of particle size and particle charge magnitude on PSL collection efficiency could be less after accounted for the interaction. The positive correlation between the drop charge magnitude and collection efficiency, and the negative correlation between the drop size and collection efficiency could be significantly less when the spray surface tension increase, as shown in Table 5.7. This model can only explain about 16% of the variance of PSL charge-specific collection efficiency. The best fit model for coal dust included the impact of particle size, particle charge magnitude, drop charge magnitude, interaction of particle size and the particle charge magnitude, and interaction of the particle charge magnitude and drop

charge on charge-specific collection efficiency, as shown in Table 5.8. There were positive correlations between particle size and the particle charge magnitude and coal dust charge-specific collection efficiency and negative correlation between the drop charge magnitude and coal dust charge-specific collection efficiency. In addition, the impact of particle size and the particle charge magnitude on coal dust collection efficiency could be less when accounting for their interaction. However, the impact of the particle charge magnitude on coal dust collection efficiency could be greater when the drop charge magnitude also increased, as shown in Table 5.8. This model can explain about 27% of the variance of coal dust charge-specific collection efficiency.

Table 5.7: Regression Coefficients and Statistical Significance Results for Best Fit Multiple Linear Regression Models of PSL Charge-Specific Collection Efficiency

Best fit Model						
Residual Standard Error =0.344, R ² =0.160, Adjusted R ² =0.151, p<0.0001, df=809		Intercept	Main effects and regression coefficients		Interaction terms and regression coefficients	
PSL charge-specific efficiency ~ Particle Size+ Particle Charge Magnitude + Particle Size: Particle Charge Magnitude+ Drop Size + Surface Tension + Drop Charge Magnitude + Surface Tension: Drop Charge Magnitude + Surface Tension: Drop Size+ Drop Size: Drop Charge Magnitude	59.1*	Particle Size***	4.62×10 ⁻¹	Particle Size: Particle Charge Magnitude*	-5.94×10 ⁻³	
		Particle Charge Magnitude***	9.05×10 ⁻³	Surface Tension: Drop Size*	6.90×10 ⁻³	
		Drop Size*	-5.14×10 ⁻¹	Surface Tension: Drop Charge Magnitude **	-2.55×10 ⁻⁵	
		Surface Tension·	-7.90×10 ⁻¹	Drop Size: Drop Charge Magnitude	-2.25×10 ⁻⁶	
		Drop Charge Magnitude**	2.05×10 ⁻³			

***p<0.001, **0.001≤p<0.01, *0.01≤p<0.05, ·0.05≤p< 0.1; otherwise, p ≥ 0.1.

Table 5.8: Regression Coefficients and Statistical Significance Results for Best Fit Multiple Linear Regression Models of Coal Dust Charge-Specific Collection Efficiency

Best fit Model						
Residual Standard Error =0.297, R ² =0.267, Adjusted R ² =0.264, p<0.0001, df=1231		Intercept	Main effects and regression coefficients		Interaction terms and regression coefficients	
Coal dust charge-specific efficiency ~ Particle Size+ Particle Charge Magnitude + Particle Size: Particle Charge Magnitude + Drop Charge Magnitude + Particle Charge Magnitude: Drop Charge Magnitude	4.65×10 ⁻²	Particle Size ^{***}	4.48×10 ⁻¹	Particle Size: Particle Charge Magnitude ^{***}	-3.79×10 ⁻³	
		Particle Charge Magnitude ^{***}	6.03×10 ⁻³	Particle Charge Magnitude: Drop Charge	3.29×10 ⁻⁷	
		Drop Charge Magnitude [*]	-1.52×10 ⁻⁵	Charge Magnitude ^{**}		

***p<0.001, **0.001≤p<0.01, *0.01≤p<0.05; otherwise, p ≥ 0.1.

5.5 Discussion

Particle diameter was the most dominant factor impacting both overall spray collection efficiency and the charge-specific collection efficiency for PSL particles and coal dust. Significantly higher capture efficiencies were observed among larger particles regardless of particle and drop charge levels (Figure 5.1), implying that the respirable particle capture tested in this study was dominated by inertial impaction and interception rather than electrical effects.

The univariate regression line suggested that the increase of particle size caused a greater increase in overall collection efficiency among smaller particles than larger particles, as shown in Figure 5.1. The collection efficiencies for particles $\geq 2 \mu\text{m}$ were about 90%, therefore collection efficiency for particles with smaller sizes were those need to be improved.

Particle charge level had a significant impact on collection efficiency. In general, an increase of 1 charge per particle could result in about 0.6-0.9% increase in spray collection efficiency (Tables 5.7 and 5.8). Although the tested surfactant-containing sprays carried either net negative charge or net positive charges, they all had greater performance on removing highly charged particles regardless the sign of charge on particles. This observation was explained by Tinsley *et al.* [72] who indicated that the collision rate between drops and particles tends to increase with increasing particle charge (irrespective of the sign of droplet charge) owing to image forces between the particles and droplets. Therefore the particle charge magnitude could be more important than the sign of particle charge in influencing particle capture efficiency. This observation may also be true for highly charged drops.

Both spray surface tension and drop size could have an impact on spray collection efficiency. Multiple regressions concluded that a decrease in spray surface tension

and drop size can cause an increase in spray collection efficiency after controlling for differences in other drop parameters (Tables 5.6 and 5.7). However, the magnitudes of these impacts were not consistent among PSL and coal dust aerosol, and there were considerable correlations among spray surface tension, drop size and drop charge. For example, even when the increase in drop size decreased spray efficiency, or when the increase in drop charge increased spray efficiency, those effects could be smaller if spray surface tension increases. Therefore, the impact of spray type itself may mask the mechanism that resulted in the changes in dust capture.

The magnitude of drop charge significantly impacts spray collection efficiency (Tables 5.5, 5.7 and 5.8). However, its impact was more consistent among PSL particles than among coal dust particles. According to the previously measured charge distributions of PSL particles and coal dust (discussed in Chapter 3), coal dust particles, especially smaller ones, had fewer charged particles than PSL particles did. Therefore, the magnitude of particle charge may play an important role on how particles interact with charged spray drops. The observation for coal dust charge-specific collection efficiency as shown in Table 5.8 also suggested that the impact of drop charge magnitude on spray efficiency can be increased if the particle charge magnitude is increased. The effect of drop charge was more important for charge-specific collection efficiency than for overall collection efficiency because the principle of electrical effects on particle capture is based on the electrical force between highly charged particles and drops. The impact of the sign of charge of the drops was less important compared to the magnitude of charge when considering electrical effect. In Chapters 2 and 3, the surfactant-containing sprays showed better collection performance on highly charged particles with opposite charges. However, the results here show that charge magnitude is more important than charge sign in influencing the collision rate between charged drops and particles, as discussed previously.

The multiple regression models for estimating spray collection efficiency may not be reliable. Even the best fit models only accounted for less than 50% collection efficiency-associated variability. There were several reasons that may cause this unreliability. First, particle size was the most dominant factor impacting spray efficiency, and the relationship between particle size and spray collection efficiency was not linear. Therefore, a linear regression cannot give a very good estimate. The univariate regression line as shown in Figure 5.1 indicates that the relationship between spray collection efficiency and particle size is similar to the relationship between single drop collection efficiency and particle size. The latter can be predicted based on the sum of three primary mechanisms, the diffusive collection efficiency, the impaction collection efficiency, and the collection efficiency due to interception [73]. However, the estimate of spray collection efficiency based on these mechanisms is complicated and cannot be described as a simple equation. The objective for this study is to investigate the impact of drop charge on spray collection efficiency when other factors were accounted for, rather than to create a predict model for spray collection efficiency. Therefore, the linear regression was used here only to control the impact of other factors. Second, the drop parameters were not well controlled in this study. The impacts of spray solution surface tension, drop size, and the drop charge magnitude were only based on the average values of each surfactant type, and did not account for differences measured in each test. Even though drop parameters were treated as continuous variables in regression models, they only had a limited number of levels (7 for PSL particle and 4 for coal dust). Thus, the predictive power of the regression models is limited by the availability of data. However, the objective of this study was not to create an accurate prediction model for spray collection efficiency. Instead, the objective is to determine if the electrical effects caused by surfactant addition to spray water can affect spray collection on charged particles, which the results of this study

are able to show. Last but not the least, variables that were not quantified by this study may also have had important effects and require future investigation.

5.6 Conclusion

Particle diameter is the most important factor that impacts respirable particle capture by surfactant spray. Significant increases in both overall collection efficiency and charge-specific collection efficiency are associated with larger size particles. Both spray surface tension and drop size impact respirable particle capture. Significant increases in collection efficiency tended to be associated with lower surface tension and smaller drop size. However, surface tension, drop size and drop charge are interrelated.

The drop charge magnitude significantly affected spray efficiency even after drop size and surface tension were taken into account. Significant increases in spray collection efficiency tend to be associated with increases in drop charge magnitude. For weakly charged particles, in addition to the magnitudes of particle and drop charge, their interaction also had important impacts on charge-specific collection efficiency for respirable particle capture. Therefore, using surfactant-containing spray does cause electrical effects that impact respirable particle capture.

Chapter 6

Conclusions and Recommendations

6.1 Overall Conclusions and Future Directions

Of the factors studied in this dissertation, particle diameter has the largest impact on respirable particle capture by water spray. Within the measured respirable dust size range in this study, the spray collection efficiency increases from 28% to 90% for coal dust when the particle diameter is increased from 0.58 μm to 3.3 μm .

Surfactant type also significantly impacts the collection of respirable dust by aqueous sprays. Adding surfactant can cause changes in spray solution surface tension, drop size and drop charge. Low spray solution surface tension and small drop size are associated with high spray collection efficiency. The sign and especially the magnitude of drop charge are also important influencers of spray collection efficiency owing to the electrical effects between dust particles and spray drops. The following findings are observed based on electrical effects: 1) The electrical effects caused by adding surfactants to spray water significantly influence spray collection efficiency. 2) Large drop charge magnitude and opposite signs of charge between dust and drops are correlated with high spray collection efficiency. 3) The sign of drop charge tends

to be associated with surfactant classification. This is not always the case, however. For example, nonionic surfactant-containing spray may carry more positive charges than cationic surfactant-containing spray. 4) The magnitude of spray drop charge is independent from the concentration of surfactant in the spray water (i.e., increasing surfactant concentration does not necessarily increase the charge level). 5) Large particle charge magnitude is also associated with high spray collection efficiency. The impact of drop charge magnitude is more significant among particles with high charge magnitude.

Tests using monodisperse PSL particles give similar results for overall collection efficiency as tests using polydisperse coal dust, but give different results for charge-specific collection efficiencies. This suggests that monodisperse PSL particles can be used as a substitute for coal dust in spray efficiency testing, but owing to different electrical properties between PSL particles and coal dust, one needs to be cautious when using PSL as a substitute for coal dust in tests that consider electrical effects.

The results have identified three areas for future study: 1) The design of the drop measurement system could be refined to improve the accuracy of drop charge distribution measurements; 2) Additional surfactants with different charge properties could be tested; 3) The impact of other drop charge parameters such as surface tension and drop size could be better controlled for.

The spray drop charge measurement system designed and built in this study can be used to measure spray drop electrostatic charge. However, an increase in length of the settling chamber, more electrical plate length options, and additional measurements to make corrections to the Drop Penetration Fraction estimate could improve the accuracy of the drop charge measurement, which is essential information for investigating the impact of drop charge on spray collection efficiency. In addition, the results here have shown that the changes in drop parameters, especially the magnitude

of drop charge, caused by adding surfactants causes the changes in spray collection efficiency. However, the initial selection of surfactants for use in this study was based on the surfactant classification and concentration, not the values of drop parameters. Therefore, different surfactants that cause different changes in drop charge properties could be useful for further investigation. Finally, in order to investigate the impact of drop charge on spray collection efficiency, other drop parameters were controlled for in this study. Only averaged values of spray solution surface tension and drop size were obtained for each surfactant type, and did not account for differences measured in each test. Therefore, a better control for surface tension and drop size in future studies could lead to a better estimate of the impact of drop parameters.

6.2 Recommendations for Surfactant Use

6.2.1 When to Use a Surfactant

In general, adding surfactants to improve spray collection efficiency for airborne dust suppression may be most effective for coal mines that have substantial numbers of particles smaller than 2 μm in diameter. These small respirable dust particles also cause the most adverse health effects. For those coal mines that mainly have larger respirable particles, adding surfactants may only have a limited benefit for airborne dust suppression. In addition, for coal mines that tend to have highly-charged coal dust, adding surfactants that are able to enhance the electrostatic charge of water drops may result in a better dust suppression.

6.2.2 How to Select a Surfactant

Surfactant charge properties are important. Surfactants that are able to cause the largest increases in water drop electrostatic charge give the largest efficiency improvements. Although the surfactant-containing sprays collect highly-charged particles of the opposite sign most efficiently, drop charge magnitude plays a more important role than the sign of drop charge in improving collection efficiency. In addition, surfactants that cause the largest reductions in spray solution surface tension or drop size may give the largest efficiency improvements.

Based on the surfactants tested here, Triton X-100 may be the best option for enhancing charged dust control by water spray owing to its ability to reduce spray solution surface tension and to carry a relatively high level of electrical charge. Adding 10^{-4} M Triton X-100 into spray water could improve collection efficiency about 0.2% on respirable coal dust with or without electrical charges. For charged respirable coal dust, Triton X-100 could improve collection efficiency by 4.5%. The ability of Triton X-100 to improve spray collection efficiency could be more significant for highly-charged aerosols.

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Appendix A

Python code for theoretical drop penetration fraction calculation used in Chapter 4

```
package main

import (
    //      "code.google.com/p/plotinum/plot"
    //      "code.google.com/p/plotinum/plotter"
    //      "image/color"
    "encoding/json"
    "fmt"
    "math"
    "net/http"
    _ "net/http/pprof"
    "os"
    "runtime"
    "time"
)

const (
    // Physical Constants //////////////////////////////////////
```

```

mug = 0.0000181 // Pa/s
rhoP = 1000.    //kg/m^3
rhoG = 1.2      //kg/m^3
g    = 9.81     //m/s^2

// Experimental Variables //////////////////////////////////////
d = 0.014 //m (plate distance)
d1 = 0.006 //m (inlet width)
d2 = 0.007 //m (outlet diameter)
//L = 0.1 //m (length of plates)
L = 0.4 //m (length of plates)

// Computational Parameters //////////////////////////////////////
deltaY = 0.000005 // m (y distance between steps)

nProcessors = 8
)

var dpBins = []float64{22.03e-6, 26.50e-6, 31.88e-6, 38.36e-6, 46.14e-6,
55.51e-6, 66.78e-6, 80.34e-6,
96.65e-6, 116.28e-6, 139.88e-6, 168.28e-6, 202.45e-6, 243.55e-6, 293.00e-6}

var Uvals = []float64{-8000, -6000, -5000, -4000, -3000, -2000, -1500,
-1000, -800, -600, -500, -400, -300, -200, -100,
100, 200, 300, 400, 500, 600, 800, 1000, 1500,
2000, 3000, 4000, 5000, 6000, 8000}

var Nvals []float64

func init() {
    const (
        // charges for 10cm
        //nmin = 1.    // minimum number of charges on droplet
        //nmax = 150000. // max charges on droplet
        // charges for 40cm
        nmin = 500    // minimum number of charges on droplet
        nmax = 400000. // max charges on droplet
        nN    = 15.
    )
}

```



```

i := 0.
numIterations := 0
for in := 0; in < len(Nvals); in++ {
    n := Nvals[in]
    nArray[in] = n
    for _, dp := range dpBins {
        for _, U := range Uvals {
            m := new(msg)
            m.dp = dp
            m.n = n
            m.u = U
            inChan <- m
            numIterations++
            i++
            fracDone := i / iTot * 100
            timeLeft := time.Duration(float64(
                time.Now().Sub(start)) *
                (100 - fracDone) / fracDone)
            fmt.Printf("\r%3.0f%% complete ``+
                ``(%v remaining)",
                fracDone, timeLeft)
        }
    }
}
// Wait for the rest of the results
for data := range outChan {
    in := getin(data.n)
    idp := getidp(data.dp)
    iu := getiu(data.u)
    fracPenetrated[iu][idp][in] = data.fracPenetrated
    numIterations--
    if numIterations == 0 {
        break
    }
}

//PlotPenetration(fracPenetrated)
out := new(outdata)
out.N = nArray

```

```

    out.Dp = dpBins
    out.U = Uvals
    out.P = fracPenetrated
    o, err := json.Marshal(out)
    if err != nil {
        panic(err)
    }
    //f, err := os.Create("results10cm.json")
    f, err := os.Create("results40cm.json")
    if err != nil {
        panic(err)
    }
    _, err = f.Write(o)
    if err != nil {
        panic(err)
    }

    elapsed := time.Since(start)
    fmt.Printf("\nProgram completed in %.3v minutes.\n",
elapsed.Minutes())
}

type msg struct {
    dp          float64
    n           float64
    u           float64
    fracPenetrated float64
}

type outdata struct {
    N []float64
    Dp []float64
    U []float64
    P [][]float64
}

func Calculate(inChan, outChan chan *msg) {
    // a constant
    for data := range inChan {

```

```

if data.n*data.u > 0. {
    // If charge and voltage are the same sign, then
    // drops are repulsed so the penetration efficiency is zero.
    data.fracPenetrated = 0.
    outChan <- data
    continue
}

e := 1.6 * math.Pow(10, -19.) //C

q := DropletCharge(data.n, e)
E := FieldStrength(data.u, d)

// Initial conditions //////////////////////////////////////
x := 0.
Ve := 0. // m/s (x-direction Electrostatic velocity)
y := 0.
// Initial guesses for other variables //////////////////////////////////
Vs := 1. // m/s (y-direction Gravitational Settling velocity)
CD := 1.
Re := 1.
for {
    Re, Vs, CD = calcCD(rhop, data.dp, g, mug, rhog,
Ve, Vs, CD, Re)

    a := Acceleration(q, E, data.dp, rhop, CD, rhog, Ve)
    deltat := deltaY / Vs
    Ve = Ve + a*deltat
    x = x + Ve*deltat
    if y >= L {
        break
    }
    y += deltaY
}
if x <= d1 {
    data.fracPenetrated = x / d1
} else if x > d1 && x <= d2 {
    data.fracPenetrated = float64(1)
} else if x > d2 && x <= d1+d2 {
    data.fracPenetrated = -1/d1*x + d2/d1 + 1
}

```

```

    } else {
        data.fracPenetrated = 0.
    }
    outChan <- data
    //fmt.Println("dp=", data.dp, "n=", data.n, "P=", data.fracPenetrated)
}
return
}

func Acceleration(q float64, E float64, dp float64, rhop float64,
    CD float64, rhog float64, Ve float64) (a float64) {
    a = (-6.*q*E)/(math.Pi*math.Pow(dp, 3.)*rhop) -
(3.*CD*rhog*math.Pow(Ve, 2.))/
    (4.*dp*rhop)
    return
}

func DragCoefficient(Re float64) (CD float64) {

    if Re <= 0.95 {
        CD = 24. / Re
    }

    if Re > 0.95 && Re <= 1.05 {
        FracStokes := (1.05 - Re) / 0.1
        FracTransition := 1 - FracStokes
        CD = 24./Re*FracStokes + 24./Re*
(1.+0.15*math.Pow(Re, 0.687))*
        FracTransition
    }

    if Re > 1.05 && Re <= 1000 {
        CD = 24. / Re * (1. + 0.15*math.Pow(Re, 0.687))
    }

    if Re > 1000. {
        e := fmt.Errorf("Reynolds number %v is too large!\n", Re)
        panic(e)
    }
}

```



```

        return
    }

    func ReynoldsNumber(dp float64, Vs float64, Ve float64, rhog float64,
        mug float64) (Re float64) {
        Re = (dp * math.Pow(Vs*Vs+Ve*Ve, 0.5) *
            rhog) / mug
        if Re > 1000 {
            fmt.Println(Vs, Ve, rhog, mug, dp, Re)
        }
        return
    }

    func SettlingVelocity(rhop float64, dp float64, g float64, mug float64,
        CD float64, rhog float64, Re float64) (Vs float64) {

        if Re <= 0.95 {
            Vs = (rhog * dp * dp * g) / (18. * mug)
        }

        if Re > 0.95 && Re <= 1.05 {
            FracStokes := (1.05 - Re) / 0.1
            FracTransition := 1 - FracStokes
            Vs = (rhog*dp*dp*g)/(18.*mug)*FracStokes +
                math.Pow(((4.*rhog*dp*g)/(3.*CD*rhog)), 0.5)*
FracTransition
        }

        if Re > 1.05 && Re <= 1000 {
            Vs = math.Pow(((4. * rhog * dp * g) / (3. * CD * rhog)), 0.5)
        }

        if Re > 1000. {
            e := fmt.Errorf("Reynolds number %v is too large!\n", Re)
            panic(e)
        }
        return
    }
}

```

```

func FieldStrength(U float64, d float64) (E float64) {
    E = U / d
    return
}

func DropletCharge(n float64, e float64) (q float64) {
    q = n * e
    return
}

func CheckConverge(n1 float64, n2 float64) (ans bool) {
    if math.Abs(n1-n2)/(n1+n2)*2 < 0.01 {
        ans = true
    } else {
        ans = false
    }
    return
}

func calcCD(rhop float64, dp float64, g float64, mug float64, rhog float64,
    Ve float64, VsIn float64, CDin float64, ReIn float64) (
    Re float64, Vs float64, CD float64) {
    i := 0
    for {
        Re = ReynoldsNumber(dp, VsIn, Ve, rhog, mug)
        Vs = SettlingVelocity(rhop, dp, g, mug, CDin, rhog, Re)
        CD = DragCoefficient(Re)
        if CheckConverge(Re, ReIn) == true &&
            CheckConverge(Vs, VsIn) == true &&
            CheckConverge(CD, CDin) == true {
            break
        } else {
            ReIn = Re
            VsIn = Vs
            CDin = CD
        }
        if i > 100 {
            fmt.Println("Cd Death Spiral!", Re, Vs, CD)
        }
    }
}

```

```

        i++
    }
    return
}

// given n, calculate array index of n
func getin(n float64) (in int) {
    for in, nx := range Nvals {
        if nx == n {
            return in
        }
    }
    panic("No matches")
    return
}

// given dp, calculate array index of dp
func getidp(dp float64) int {
    for idp, dpx := range dpBins {
        if dpx == dp {
            return idp
        }
    }
    panic("No matches")
}

func getiu(u float64) int {
    for iu, ux := range Uvals {
        if ux == u {
            return iu
        }
    }
    panic("No matches")
}

```

Appendix B

Python code for data inversion used in Chapter 4

```
import process
import numpy
import multiprocessing

plateLens = ["10 cm", "40 cm"]
Cs = [process.chargeSurface('results10cm.json'),
#      process.chargeSurface('results40cm.json')]

dpBinEdges = numpy.array([20.00, 24.06, 28.95, 34.82, 41.89, 50.40, 60.63, 72.94,
      87.75, 105.56, 126.99, 152.77, 183.79, 221.11, 266.00, 320.00]) # um
dpBinCenters = Cs[0].Dp * 1.e6

nsimulations = 100 # number of monte carlo simulations

voltages = Cs[0].U
outputFolder = "../output/"

def runTest(num, index):
```

```

testName = "Test"+str(test)
folder = "../"+testName
r = process.report(outputFolder + "dropletPlots"+testName+".tex")

process.mkdir(outputFolder + "plots"+testName)

eff = {}
std = {}

for plateLen in plateLens:
    eff[plateLen] = numpy.zeros((len(voltages),len(dpBinCenters)))
    std[plateLen] = numpy.zeros((len(voltages),len(dpBinCenters)))

    for iV,voltage in enumerate(voltages):
        if voltage > 0.: sign = "P"
        else: sign = "N"

        iData = index[plateLen][sign+str(abs(voltage))]
        fnames = process.parseFileNames(folder,plateLen,"A2",
            iData['A2before'])
        backgroundMeanBefore, backgroundStdBefore, binEdges = \
            process.groupHist(dpBinEdges,fnames=fnames)
        fnames = process.parseFileNames(folder,plateLen,"A2",
            iData['A2after'])
        backgroundMeanAfter, backgroundStdAfter, binEdges = \
            process.groupHist(dpBinEdges,fnames=fnames)

        fnames = process.parseFileNames(folder,plateLen,"A1",
            iData['A1before'])
        totalMeanBefore, totalStdBefore, binEdges = process.groupHist(\
            dpBinEdges,fnames=fnames)

        fnames = process.parseFileNames(folder,plateLen,"A1",
            iData['A1after'])
        totalMeanAfter, totalStdAfter, binEdges = process.groupHist(\
            dpBinEdges,fnames=fnames)

    frac = iData['order']

```

```

totalMean = totalMeanBefore * (1-frac) + totalMeanAfter * frac
totalStd = totalStdBefore * (1-frac) + totalStdAfter * frac
backgroundMean = backgroundMeanBefore * (1-frac) \
+ backgroundMeanAfter * frac
backgroundStd = backgroundStdBefore * (1-frac) \
+ backgroundStdAfter * frac

chargedMean, chargedStd, binEdges = process.groupHist(\
    dpBinEdges,
    fnamePattern=folder + "/" + plateLen + "/A2 "+" \
        sign+str(abs(voltage))+".vsp")

eff[plateLen][iV,:],std[plateLen][iV,:] = \
    process.calcEfficiencyAddBackground(\
        chargedMean,chargedStd,backgroundMean,backgroundStd,
        totalMean,totalStd)

histName1 = "plots"+testName + "/" +plateLen+sign+\
        str(abs(voltage))+ "_background.pdf"
histName1 = histName1.replace(" ", "")
backgroundStd = numpy.sqrt(backgroundStd**2 + totalStd**2)
process.plotHist(outputFolder+histName1,backgroundMean+totalMean,
        binEdges,std=backgroundStd, ylabel="Background count")

histName2 = "plots"+testName + "/" +plateLen+sign+\
        str(abs(voltage))+ "_charged.pdf"
histName2 = histName2.replace(" ", "")
process.plotHist(outputFolder+histName2,chargedMean,
        binEdges,std=chargedStd,ylabel="Charged count")

histName3 = "plots"+testName + "/" +plateLen+sign+\
        str(abs(voltage))+ "_eff.pdf"
histName3 = histName3.replace(" ", "")
process.plotHist(outputFolder+histName3,eff[plateLen][iV,:],
        binEdges,std=std[plateLen][iV:],
        ylabel="Efficiency (fraction)")

r.addFigure([histName1,histName2,histName3],

```

```

        plateLen + sign+str(abs(voltage)),3)

outfs = []
resultOutFs = []
for plateLen in plateLens:
    f = open(outputFolder + testName + " " + plateLen + ".csv", 'w')
    f.write("size,charge,run,frac\n")
    outfs.append(f)
    f2 = open(outputFolder + testName + " " + plateLen + " eff.csv", 'w')
    f2.write("size,model or measure, run,eff or voltage\n")
    resultOutFs.append(f2)

for idp,dp in enumerate(dpBinCenters):
    print testName, dp
    if dp < 320 or dp > 130: #drop size range for 40 cm plates
        continue
    for plateLen,c,f,f2 in zip(plateLens,Cs,outfs,resultOutFs):
        charges, modelVoltages, modeleff, coeffs,modelFit = \
            c.chargeFit(idp,
                eff[plateLen][:,idp],std[plateLen][:,idp], nsimulations)

        for ii in range(coeffs.shape[0]):
            for jj in range(coeffs.shape[1]):
                f.write("%g,%g,%g,%g\n"%(dp,charges[jj],
                    ii,coeffs[ii,jj]))

        f2.write("%g,measured (mean),xx"%(dp))
        for val in eff[plateLen][:,idp]:
            f2.write(",%g"%val)
        f2.write("\n")
        f2.write("%g,measured (std),xx"%(dp))
        for val in std[plateLen][:,idp]:
            f2.write(",%g"%val)
        f2.write("\n")
        f2.write("%g,model voltages,xx"%(dp))
        for val in modelVoltages:
            f2.write(",%g"%val)
        f2.write("\n")
        for ii in range(coeffs.shape[0]):

```

```

        f2.write("%g,model,%i"%(dp,ii))
        for val in modelFit[ii,:]:
            f2.write(",%g"%val)
        f2.write("\n")

        fname1 = "plots"+testName + "/" + plateLen + str(int(dp)) \
+ "um_chargeDistribution.pdf"
        fname1 = fname1.replace(" ", "")
        process.plotCharges(outputFolder+fname1, charges, coeffs)
        fname2 = "plots"+testName + "/" + plateLen + str(int(dp)) \
+ "um_chargeDistributionCounts.pdf"
        fname2 = fname2.replace(" ", "")
        process.plotChargeCounts(outputFolder \
+ fname2, charges, coeffs, totalMean, idp)
        fname3 = "plots"+testName + "/" \
+ plateLen + str(int(dp)) + "um_eff_vs_voltage.pdf"
        fname3 = fname3.replace(" ", "")
        process.plotEfficiency(c, outputFolder+fname3,
            eff[plateLen][:, idp], std[plateLen][:, idp], modeLeff, modelFit,
            voltages/1000., modelVoltages/1000., dp*1.e-6, "Voltage (kV)")
        r.addFigure([fname1, fname2, fname3],
            plateLen + str(dp) + " $\\mu$m")

    r.write()

if __name__ == '__main__':
    index = process.readIndex("../DataIndex.xlsx")
    jobs = []
    for test in range(1,8):
        p = multiprocessing.Process(target=runTest, args=(test, index[test]))
        jobs.append(p)
        p.start()

```