We would like to thank the reviewers for their helpful comments and suggestions. We have addressed each of their comments below and made appropriate changes in the paper.

Reviewer 1 comments:

COMMENT:

P.4 line 138 and following: the model description is cryptic and confusing, with the same symbols used for the mean values and standard deviations of three different quantities, diameter, charge density and initial vertical velocity. E.g. the variable described first as the standard deviation of the size distribution has the dimension of surface charge density (line 187). This makes it unnecessarily difficult to assess the methodology.

RESPONSE:

We have distinguished the different means and standard deviations that are associated with the three parameters. This change is seen in lines 137-139.

COMMENT:

Another potential confusion occurs in section 5: on p.7 lines 211-212 it is stated that the electrostatic forces were calculated as the product of the electric field and the surface charge density, meant here to be the same charge density as that on the particle surfaces. Taken at face value, this might be understood to imply that the atmospheric system is treated as a parallel capacitor, with the charge density equal to that found on particle surfaces (or on bulk quartz surfaces, as in one cited reference, Miura and Arakawa, 2007). This is misleading at best, and could potentially hide a basic error made by the authors - which is presumably not the case, judging from equation 2, which is correct.

Having said that, there is some justification for employing the field-density product, as both these parameters are unknown and the electrostatic force does depend on their product. So it should be explained clearly what is meant.

RESPONSE:

We have clarified the definition and usage of σE in lines 210 to 232.

COMMENT:

My main objection concerns the discussion of the results. It is concluded that even the fair weather electric could maintain large dust particles aloft. However, this is assuming extreme value of surface charge densities, 220 μCm^{-2}, found in lab studies of bulk quartz surfaces (Miura and Arakawa, 2007), and untypically large (compare e.g. the values cited in Angus and Greber, 2018). More realistic values, especially for particulate matter rather than bulk material, should be cited from literature (which is not as sparse as the discussion implies, if spatial charge density measurements are included) and used in these conclusions. The authors own lab results point to charge densities two orders of magnitude lower than the above number, and similar values were found by Waitukaitis et al. (2014). So it should be concluded that electric fields exceeding the normal fair weather value are probably needed, as suggested previously by some authors. For example, it was estimated on the assumption of realistic spatial charge densities that fields of 2 kV/m would be required to maintain dust in suspension (Ulanowski et al., 2007). While such high values exceed the fair weather field, extensive literature suggests that they are not uncommon during dust episodes.

RESPONSE:

First, we have dialed-down our claims of atmospheric implications. For example, we replaced the sentence "Thus, our experimental and modelling results indicate that electrostatic forces should be considered when determining the effect of atmospheric dust on the climate" in the abstract with "Thus, our experimental and modelling results indicate that electrostatic forces may in some cases be relevant regarding the effect of atmospheric dust on the climate".

Second, we have rewritten the section from lines 210 to 232 and added extra detail to lines 282 to 304 to show that both the particle charge and electric field can vary substantially, and that the calculated value for σE of 38 mC V m^{-3} is on the lower part of the given range of σE. We now refer to ranges of values rather than specific values. We cite multiple well-regarded sources to justify the range of values.

COMMENT:

These unsupported conclusions are made prominent in Fig. 6 and 7, where the earlier convention to use the field-density product (as in Fig. 5) is unaccountably abandoned in favour of citing just the electric field. The hidden assumption, not mentioned in the figure captions, is the extreme, and probably unrealistic, value of the surface charge density. This is misleading, as all that can be said is that the field-density product has a certain value. So the labeling of both figures should be changed.

RESPONSE:

For figures 6 and 7, the charge density follows a distribution. We have edited the figures to show the standard deviation of σE used to perform each set of calculations with the charge density of 110 $\mu C/m^2$. As described above, we do not believe these values to be unrealistic. We have made edits in the text (lines 251 to 262 and 277) to go along with the changes to the figures.

COMMENT:

Small points and typos:

P.1 line 30: for completeness, dust also interacts via the semi-direct effect, whereby dust enhances cloud evaporation.

P. 8 line 257: presumably what is meant is that particles "would remain aloft", not "would be lofted"?

Fig. 5: the year cited should be 2003.

Fig. 7: In my view, the cumulative distribution shown is less clear than the alternative form of a differential distribution, and should be changed.

RESPONSE:

We have fixed these errors in the text, as well as checked thoroughly through to ensure that there are no more mistakes. We have also replaced figure 7 with a differential distribution rather than a cumulative distribution.

Reviewer 2 comments:

COMMENT:
I thank the authors for taking into account my previous comments and for their reply. Unfortunately, my main objection is still unanswered. The authors tried to apply the findings of their short time and small scale experiment, to the long transport of dust particles in the atmosphere. They assumed that the dust particle charge can obtain very large values, and the fair weather electric field, that can be sustained in several days, are able to create the necessary electrical force to counterbalance the gravitational force and transport the particles in large distances. I do not argue, that in the laboratory, very high values of surface charge density can appear, as a consequence of the turboelectric effect. My objection is that these high values are impossible to be met in the atmosphere. I will prove my point as follows.

The highest electric field value that can appear in the atmosphere has been measured in thunderstorms right before the initiation of a lightning flash. When the electric field exceeds a specific limit, the air is ionized that leads to the discharge of the charged particles and therefore to the reduction of the electric field. Up to now there has been no indication nor observation of lightning flashes in dust clouds. Therefore, it is a valid assumption that the electric field inside a dust cloud can never exceed the lightning initiation threshold. Let's assume this threshold to be equal to ±400 kV/m [*Stolzenburg et al.*, Geophys. Res. Lett., 34, 2007]. This threshold is scaled along the altitude as a function of the neutral density. Assuming the US Standard Atmosphere 1976, the vertical distribution of the electric field threshold is shown in Fig. 1.

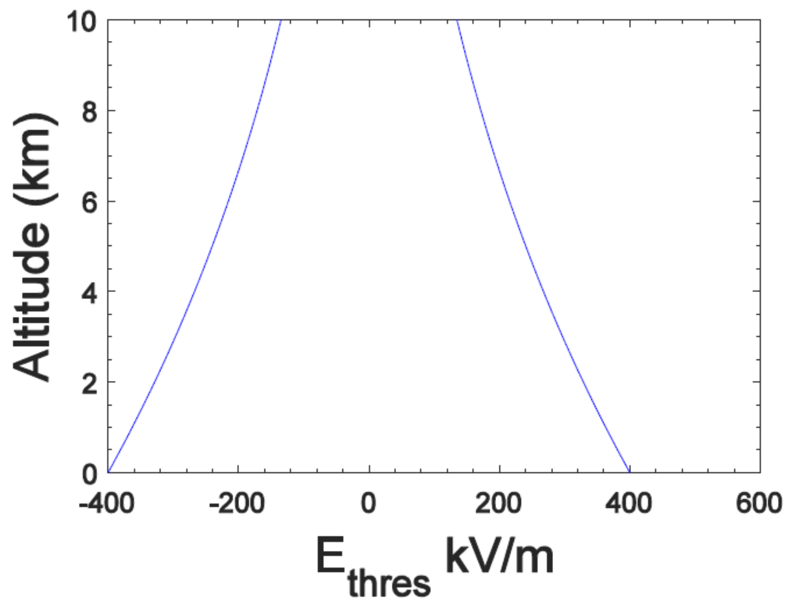

Fig. 1: Vertical distribution of the E_{thres}.

The total electric field E_{tot} at the surface of a spherical particle cannot exceed this threshold. This electric field can be written as $E_{tot} = E_{particle} + E_{ambient}$, where $E_{particle}$ is the electric field due to the charge of the particle, and $E_{ambient}$ is the ambient electric field is the large scale electric field produced by other charged particles or other mechanisms (e.g. the potential difference between the Ionosphere and the Earth's surface).

The case of a positively charged particle inside a positive electric field (pointing upwards to the

Ionosphere) is examined, but the methodology can be applied in any charge polarity and in any polarity of the electric field. Moreover, the particle is assumed as a conductor of electricity in electrostatics [*Ulanowski et al.*, Atmos. Chem. Phys., 7, 2007].

Under these valid assumptions, the surface charge density $\sigma_{particle}$ of the particle, can never exceed the value $\sigma_{lim}=(E_{thres}-E_{ambient})\varepsilon_0$, where ε_0 is the vacuum permittivity. Fig. 2 and Fig.3 show the vertical distribution of the σ_{lim} for positively and negatively charged particles, respectively. It is noted, that since an upward electric field is assumed, the electrical force for the positively charged particles tends to counterbalance the gravitational force, while the electrical force for the negatively charged particles acts in the same direction with the gravitational force leading to the faster fall of the particles (for an opposite polarity electric field obviously the opposite conclusion holds).

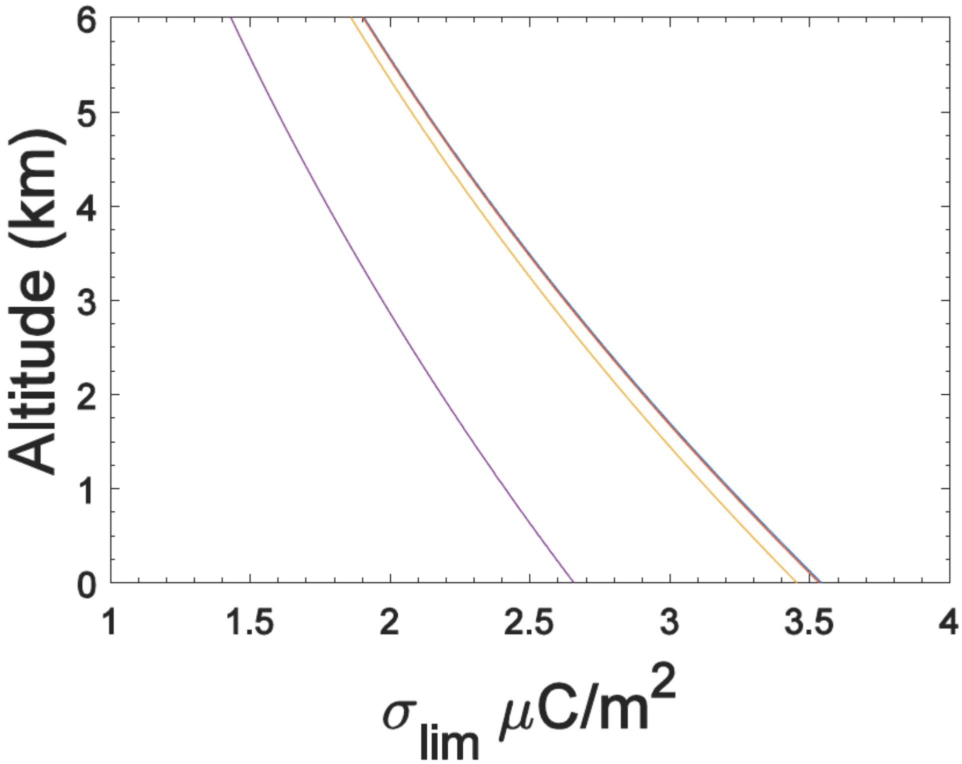

Fig.2: Vertical distribution of σ_{lim} for positively charged dust particles.

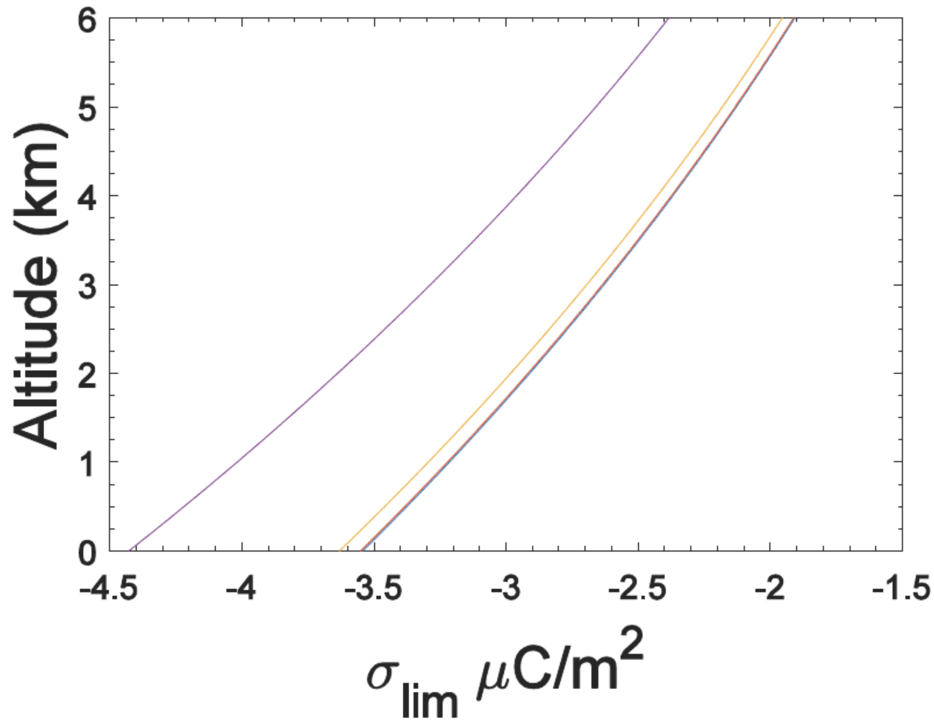

Fig.3: Vertical distribution of σ_{lim} for negatively charged dust particles.

In Fig. 2 and Fig. 3, the blue line corresponds to $E_{ambient}$ = 100 V/m, the red line corresponds to $E_{ambient}$ = 1000 V/m, the orange line corresponds to $E_{ambient}$ = 10 kV/m, and the purple line corresponds to $E_{ambient}$ = 100 kV/m.

If we are interested in the case that the electrical force counteracts the gravitational force, it is clear from Fig. 2 that the surface charge density is physically impossible to exceed the value of 3.5-3.6 $\mu C/m^2$. This is two orders of magnitude lower than the assumed upper limit value of the authors. Therefore, in their analysis in lines 230-240, the normal distribution of the surface charge density that has to be assumed for the long range transport, must have standard deviation s=3.5/3 $\mu C/m^2$, because 3s=3.5 $\mu C/m^2$. By doing that, the authors will realize that much stronger electric fields are required than the fair weather electric field, for the long range transport of the dust particles. Thus, the discussion section has to be modified accordingly.

RESPONSE:

Thank you for the comments. We agree with the reviewer that the threshold charge density on a particle is limited by the threshold electric field from the particle that would cause a discharge into air. However, a complete analysis of this effect shows that the magnitude of particle charges we discuss in the paper in fact leads to electric fields below this threshold. Here, we expanded on the reviewer's back-of-the-envelope calculations to find the maximum surface charge density on particles based on Paschen's law, which describes the magnitude of the electric field threshold for gas breakdown.

The electric field from the particle that leads to gas discharge, E_t, is given by Paschen's law, which is a function of the distance from the particle surface and the ambient pressure,[4–7]

$$E_t = \frac{Bp}{\ln\left(\frac{A}{ln\left(\frac{1}{\gamma}+1\right)}\right) + \ln(pd)} \qquad (1)$$

where p is the ambient pressure, d is the distance from the surface, and A, B and γ are constants equal to 112.5 V $(\mathrm{kPa\ cm})^{-1}$, 2737.5 $(\mathrm{kPa\ cm})^{-1}$, and 0.01 respectively.[7]

The key point underlying Paschen's law is that gas breakdown occurs by an electron avalanche, wherein free electrons are accelerated by the electric field and undergo collisions with other molecules that release more free electrons, and each of these newly freed electrons are accelerated and collide to form even more free electrons, and so on.[8–11] In order for an electron avalanche to occur, the electric field must operate over a sufficiently long distance so as to enable a series of collisions to enable the avalanche to develop. For this reason, the threshold electric field diverges to infinity at small gap sizes.

We now address the maximum charge on for a spherical particle, as limited by Paschen's law. Importantly, we note that the electric field from the particle decreases as a function of distance from the particle surface, d, as given by Gauss's Law,

$$E = \frac{q}{4\pi\epsilon(R+d)^2} \qquad (2)$$

where R is the radius of the particle, q is the charge given by $q = 4\pi R^2\sigma$ where σ is the surface charge density of the particle. The maximum charge density possible before gas breakdown occurs when the electric field from the particle (Eq. 2) becomes larger than the electric field threshold from Paschen's law[8,11] (Eq. 1), noting that both of these quantities depend on the distance d from the particle surface. As depicted in Figure 4, for a 10 µm particle, the maximum charge density prior to gas breakdown is 2800 $\mu C/m^2$; the maximum charge density on a particle is dependent on the particle size.

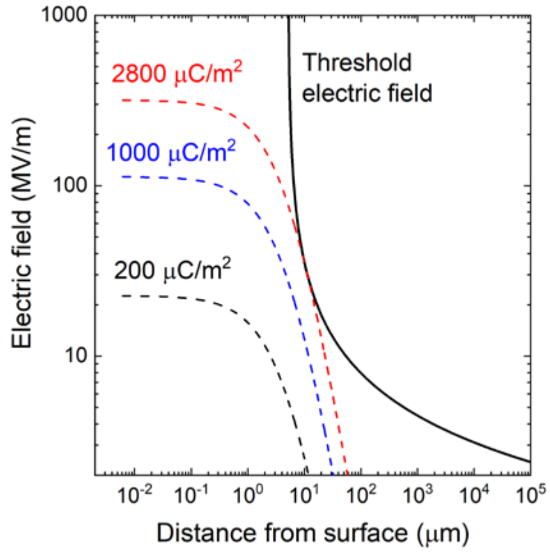

Figure 4. Electric field due to a 10 μm particle with different charge densities (dashed lines) and the threshold electric field given by Paschen's law (solid line)

Using this rigorous criterion, we obtain the maximum charge density for different particle sizes at a pressure of 54 kPa, characteristic of 5 km altitudes. As shown in Figure 5, see the maximum surface charge density is higher than 200 μC/m^2 at all relevant particle sizes.

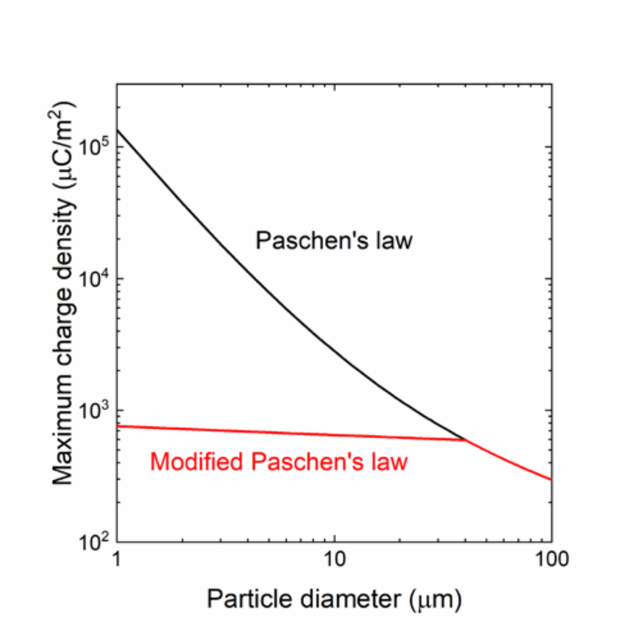

Figure 5. Maximum surface charge density for different sized particles limited by gas breakdown described by Paschen's law (black) and Modified Paschen's law (red).

We note that at very small distances, recent work has shown that the breakdown electric field is more accurately represented by a Modified Paschen's law, which takes into account field emission.[5,6,11] As shown in Figure 5, the conclusions described above, regarding charge densities on the order of 200 μC/m^2, hold even when the Modifield Paschen's law is used in the analysis. The charge densities on surfaces of this order of magnitude and higher is supported by several experiments.[2,10,12–14] Thus, particle charge densities exceeding 200 μC/m^2 are physically possible.

We added details in lines 210 to 231 and lines 282 to 304.

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Electrostatic forces alter particle size distributions in atmospheric dust

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Abstract. Large amounts of dust are lofted into the atmosphere from arid regions of the world before being transported up to thousands of kilometers. This atmospheric dust interacts with solar radiation causing changes in the climate, with larger-sized particles having a heating effect, and smaller-sized particles having a cooling effect. Previous studies on the long-range transport of dust have found larger particles than expected, without a model to explain their transport. Here, we investigate the effect of electric fields on lofted airborne dust by blowing sand through a vertically-oriented electric field, and characterizing the size distribution as a function of height. We also model this system, considering the gravitational, drag, and electrostatic forces on particles, to understand the effects of the electric field. Our results indicate that electric fields keep particles suspended at higher elevations and enrich the concentration of larger particles at higher elevations. We extend our model from the small-scale system to long-range atmospheric dust transport to develop insights on the effects of electric fields on size distributions of lofted dust in the atmosphere. We show that the presence of electric fields and the resulting electrostatic force on charged particles can help explain the transport of unexpectedly larger particles and cause the size distribution to become more uniform as a function of elevation. Thus, our experimental and modelling results indicate that electrostatic forces may in some cases be relevant regarding the effect of atmospheric dust on the climateThus, our experimental and modelling results indicate that electrostatic forces should be considered when determining the effect of atmospheric dust on the climate.

1 Introduction

The global climate is largely governed by a radiative balance between incoming solar and outgoing thermal radiation from the Earth (Hansen, 2005). This balance depends on interactions between matter and radiation. Dust represents an increasingly significant amount of matter in the atmosphere and interacts with radiation both directly, through scattering and absorption (Tegen, 2003; Kok et al., 2018) and indirectly, by impacting processes such as cloud formation and dissipation and biogeochemical feedbacks (Mahowald, 2011). The source of this dust is typically arid regions such as northern Africa and central Asia, where wind ejects dust into the atmosphere (Bagnold, 1971; Kok et al., 2012). Atmospheric dust can travel far from its source (Betzer et al., 1988) – e.g., Saharan dust travels to the western hemisphere – and so dust events in one region can affect climate in distant regions. Anthropogenic factors such as over-cultivation and water diversion significantly increase the amount of atmospheric dust (Ginoux et al., 2012; Webb and Pierre, 2018). Thus, accurately characterizing the effects of atmospheric dust on the Earth's radiative balance is necessary for understanding the anthropogenic role in climate change.

The size distribution of dust systems has been shown to have important implications on the climate (Mahowald et al., 2014). Scattering and absorption of different wavelengths of light depend on the size distribution of airborne particles (Tegen and Lacis, 1996). Smaller-sized particles predominately scatter solar radiation, which has shorter wavelengths, away from the Earth, causing a cooling effect on the climate (Kok et al., 2017). In contrast, larger-sized particles predominately scatter thermal radiation emitted by the Earth, which has longer wavelengths, back towards the earth, causing a heating effect on the climate. In addition, larger particles are more effective at absorbing both solar radiation and thermal radiation from the earth, trapping heat in the atmosphere (Otto et al., 2007). The net effect of atmospheric dust on the climate thus depends on the particle size distribution of the dust.

Previous field studies have found surprising results regarding the particle size distributions of dust transported far from the source (Betzer et al., 1998; Maring et al., 2003; Reid et al., 2003; Haarig et al., 2017; Renard et al., 2018; van der Does et al., 2018). The distributions contain larger particles than predicted by models, which, as described above, could have implications on the climate. Various mechanisms could be responsible for this effect including electrostatic forces (Ulanowski et al., 2007; van der Does et al., 2018).

It has been known for over 100 years that dust particles can become highly charged, as indirectly observed by the presence of strong electric fields in dust storms (Rudge, 1913). These electric fields can be large, with magnitudes greater than 100 kV m^{-1} (Schmidt et al., 1998). The electric fields develop as follows. Wind-blown dust particles collide with one another and transfer charge by triboelectric charging; this process is the same as that occurring when a balloon is rubbed on hair. Triboelectric charging occurs even when all particles are composed of the same material, with some particles charging positive and others charging negative (Lacks and Sankaran, 2011; Xie et al., 2013). Due to the cancelling contributions of positive and negative particles, the volumetric charge density in lofted dust systems is expected to be very small, while the charge on each individual particle can still be large. Studies have shown that, to some extent, smaller and larger particles tend to charge with different polarities (Zhao et al., 2002; Forward et al., 2009; Bilici et al., 2014; Waitukaitis et al., 2014; Toth et al., 2017). This particle-size-dependent polarity of charge, combined with the separation of small and large particles in a gravitation field such that smaller particles are lofted higher, generates the electric fields in dust storms.

The electric fields generated in dust storms have been shown to cause dust liftoff, i.e., transferring particles from the ground to the air (Kok and Renno, 2006, 2008). The dust liftoff was subsequently confirmed by wind tunnel experiments (Rasmussen et al., 2009). Recently, field studies of dust storms found a strong correlation between the electric field and the density of airborne particles (Esposito et al., 2016), which supports the prediction that electric fields lead to dust liftoff.

In this paper, we address the effects of electrostatics on lofted dust, i.e. particles that are already airborne. The electric fields leading to these effects could be created in dust storms, as described above, or could occur in the atmosphere for other reasons (e.g., fair weather electric fields). The effect of electric fields on airborne dust are fundamentally different than those on dust liftoff, where physical contact with the ground surface allows charge transfer from the particles to (electrical) ground. To reveal how electric fields affect airborne dust, we carried out controlled laboratory experiments in which electric fields are applied to lofted particles and characterized the particle size distributions as a function of the lofted height. We also develop a model to support our experimental results, and then apply the model to prior studies to elucidate the role of electric field on long-range dust transport.

2 Experimental Methods

We constructed a setup to characterize the effects of an electric field on dust trajectories after lift-off. The setup, shown in Fig. 1, consisted of two parallel electrodes (89 cm long, 15 cm wide) connected to a DC high-voltage power supply (HB-Z303-1AC) oriented such that the electric field between the electrodes was perpendicular to the ground surface. The distance between the electrodes was 12 cm, and the electric field was varied from 125 to -125 kV m^{-1} where the polarity of the electric field was defined by the polarity of the top electrode; i.e., a positive electric field had a positive top electrode and a negative electric field had a negative top electrode. In order to prevent particle contacts with the electrodes and ensure that the sand particles contacted only other sand particles (i.e., not other surfaces), the electrodes were covered with a thin layer of sand held in place by two-sided tape.

A 250 g sand bed 1.5 cm tall, 20 cm long, and 11 cm wide was positioned 4 cm upstream of the electrodes. The sand was polydisperse and characterized by a mean diameter of ~132 µm, 40% by mass with diameters smaller than 105 µm, and 60% by mass with diameters between 105 and 450 µm. Particles with diameters greater than 450 µm were removed by sieving. A fan placed 15 cm upstream of the sand bed was used to blow sand through the electrodes. The average air speed over the sand bed was 6.7 m s^{-1}, and decreased to 3.2 m s^{-1} at the end of the electrodes. As it is difficult to control exactly when particles become charged, the particles likely had an initial charge in the sand bed due to particle contacts during sample preparation and could acquire more charge during lift-off and transport between the electrodes. Each trial ran for 2 minutes, consuming approximately 80% by mass of the sand bed.

To collect particles, two cups were placed 11 cm downstream the electrodes with slit openings 2.2 cm tall and 11 cm wide. The bottom cup collected particles between heights of 0 and 2.2 cm, and the top cup collected particles between heights of 8.8 and 11 cm. The total mass collected in both cups in the experiments ranged from 8 g to 35 g, which represents approximately 21% of the mass passed through the system by the fan. The particles collected in the two cups were sieved with a 105 µm mesh sieve; "small particles" were defined as those that passed through the sieve and "large particles" were defined as those that did not pass through. After sieving the particles in each cup, we determined the mass of the small and large particles collected in each cup, m_i^j, where i designates the particle size ($i = S$ for small particles, $i = L$ for large particles) and j designates the cup position ($j = T$ for top, $j = B$ for bottom).

3 Modeling Methods

We developed a Monte Carlo model to address the effects of electric fields on dust transport in both our experimental system as well as previous field studies of dust transported long distances in the atmosphere. Our simulations address the final positions of wind-blown particles under various conditions. We assumed the particles are spheres of diameter, D, with density, ρ, and surface charge density, σ. In the horizontal direction, we do not consider forces on the particles and we assumed that the particles travel at the velocity of the wind. In the vertical direction, we assumed that the only forces acting on the particles are gravitational, electrostatic, and drag forces. The gravitational force, F_G, is given by,

$$F_G = \frac{\pi D^3}{6} \rho g \quad (1)$$

where g is the acceleration due to gravity. The electrostatic force, F_E, is given by,

$$F_E = \pi D^2 \sigma E \quad (2)$$

where E is the electric field strength. The drag force, which always acts in the direction opposite to vertical velocity, is given by,

$$F_D = -\frac{1}{8} C_D \rho_f \pi D^2 v^2 \hat{v} \quad (3)$$

where ρ_f is the density of air, v is the velocity of the particle in the vertical direction, and $\hat{v}$ is the direction of motion of the particle. We neglected air velocity in the vertical direction. For the drag coefficient, C_D, we used the following correlation (Brown and Lawler, 2003),

$$C_D = \frac{24}{Re}(1 + 0.15 Re^{0.681}) + \frac{0.407}{1 + \frac{8710}{Re}} \quad (4)$$

where Re is the Reynolds number,

$$Re = \frac{\rho_f |v| D}{\eta} \quad (5)$$

and η is the viscosity of air. We note that the drag and electrostatic forces can act upwards or downwards depending on the direction of particle velocity and the sign of σE respectively. The net force on a particle, F_{net}, is the sum of F_G, F_E, and F_D.

The equations governing the motion of the particle in the vertical direction are

$$\frac{dv}{dt} = \left(\frac{6}{\pi D^3 \rho}\right) F_{net} \quad (6)$$

and

$$\frac{dy}{dt} = v \quad (7)$$

where y is the position of the particle in the vertical direction.

Particle trajectories are determined by numerically integrating Eqns. (6) and (7). At sufficiently long times, the system attains a constant terminal velocity – i.e., as the particle velocity increases due to acceleration from gravitational and electrostatic forces, the drag force increases until it balances other forces and $F_{net} = 0$. Thus, steady-state particle trajectories can be obtained by calculating the terminal velocity, rather than by integrating the equations of motion.

Since particles traverse the electrodes more quickly (18 ms) than terminal velocity can be reached in our laboratory experiments, particle trajectories were obtained by numerical integration of the equations of motion. In the horizontal direction, particles move at the average wind speed in the system (4.95 m s^{-1}). In the vertical direction, particles start with size-dependent initial vertical velocities, at an initial height of zero. We considered a collection of particles of density, $\rho = 2600$ kg m^{-3} (characteristic of quartz), where the particle diameter, charge, and initial vertical velocity, v_0, were chosen randomly from normal distributions characterized by ~~a specified~~respective means, μ, and standard deviations, s. For particle diameter, $\mu_D = 132$ μm and $s_D = 60$ μm (as in the experiments). For surface charge density, $\mu_\sigma = 0$ and $s_\sigma = \alpha$, where α is a fitting parameter. For initial vertical velocity, $\mu_{v_0} = 0$ and $s_{v_0} =$

β/D^2 that depends on particle diameter, where β is a fitting parameter (physically, this relationship causes smaller particles to have higher initial velocities); only positive initial velocities were sampled from the distribution. Particle trajectories were calculated using Euler's method with a time step of 18 μs. After traveling the length of the electrode (0.89 m), particles with heights between 0 and 6 cm were considered to be collected in the bottom cup, and those with heights greater than 6 cm were considered to be collected in the top cup. Particles with heights less than 0 cm were not considered to be collected in the collection cups.

For long-range dust transport, particles reach terminal velocity, and thus a simpler method can be used to determine particle trajectories. The dust was assumed to be already lofted and following the model of Maring et al. (2003), particles were uniformly distributed in a vertical 2000 m window, with initial velocities equal to their terminal velocities. We considered a collection of particles of density $\rho = 2600$ kg m^{-3}, and the particle diameter and charge were chosen based on the study being modeled. The terminal velocity was found by numerically solving for the velocity at which $F_{net} = 0$. The final height of each particle after traveling for a given amount of time was obtained simply by calculating the initial height plus the terminal velocity multiplied by time.

4 Laboratory Studies

First, we address the effect of the applied electric field on the elevation of airborne particles. We quantify this effect by the fraction of total collected particles in the top cup, F^T,

$$F^T = \frac{m_L^T + m_S^T}{m_L^T + m_S^T + m_L^B + m_S^B}. \tag{9}$$

Figure 2 shows our results for F^T as a function of applied electric field. As the magnitude of the electric field increases, more of the airborne dust remains suspended higher, as indicated by the increase in the fraction of particles collected in the top cup. We note that this enhancement of dust elevation occurs with either polarity of electric field.

We now turn to the particle size dependence of the electric field effect. Figure 3(a) shows results for the fraction of large particles that are collected in the top cup rather than the bottom cup, g_L^T,

$$g_L^T = \frac{m_L^T}{m_L^T + m_L^B} \tag{10}$$

Likewise, Fig. 3(b) shows results for the fraction of small particles that are collected in the top cup rather than the bottom cup, g_S^T,

$$g_S^T = \frac{m_S^T}{m_S^T + m_S^B}. \tag{11}$$

The values g_L^T and g_S^T show the fraction of large and small particles, respectively, in the system that are collected in the top cup. When no electric field is applied, g_S^T is greater than g_L^T meaning that more of the small particles are in the top cup than of the large particles in the top cup. This is due to gravitational effects which cause smaller particles to remain suspended higher than larger particles. As the magnitude of the electric field increases, large and small particles both remain suspended higher, as shown by the increase in the fractions of large and small particles in the top cup.

We now examine how the size distribution of particles at higher elevations is affected by the electric field. The fraction of particles in the top cup that are large, f_L^T, is defined as

$$f_L^T = \frac{m_L^T}{m_L^T + m_S^T}. \tag{12}$$

While f_L^T can be obtained directly from the experimental data, these results are especially susceptible to noise, as the overall ratio of large to small particles collected varies between trials. This could be due to the fact that the initial particle size distribution could change over time as small particles are preferentially lost from the system. We can get a better estimate of f_L^T by calculating it independently, using variables that are less susceptible to noise,

$$f_L^T = \frac{g_L^T F_L}{F^T}, \tag{13}$$

where F_L, the fraction of particles in both cups that are large, is defined as

$$F_L = \frac{m_L^T + m_L^B}{m_L^T + m_S^T + m_L^B + m_S^B}, \tag{14}$$

g_L^T and F^T come from fits to our experimental data, and the average value of F_L across all trials is 0.63 ± 0.01. The results for f_L^T are shown in Fig. 4. We see that an electric field causes the particle size distribution at higher elevations to be enriched in larger particles.

We carried out simulations of our model for the experimental system. The fitting parameters were varied until the model showed a reasonable fit to the experimental results for $\alpha = 1.4$ μC m^{-2} and $\beta = 11000$ μm^2. We note that surface charge densities characterized by $\alpha = 1.4$ μC m^{-2} are reasonable, as they are similar to results found experimentally in glass particle systems (Waitukaitis et al., 2014), and they are significantly smaller than surface charge densities measured for triboelectrically charged quartz (Miura and Arakawa, 2007; Shen et al., 2016). Results were obtained for trajectories of 10^6 particles. As seen in Figs. 2-4, our model results match our experimental results reasonably well. In both our model and experiments, the electric field maintains particles at higher elevations and shifts the size distribution at higher elevations towards large particles. While the distributions for particle size and charge density are not perfectly representative of the particles in our experimental system, the trends in the model will be the same regardless of what distributions are used.

5 Modeling Field Studies

Since the effect of dust on the climate is dependent on particle size, to accurately gauge the role of atmospheric dust on the climate, the size distribution of airborne dust must be known. Our results indicate that electric fields can alter the size distribution of transported dust. As we describe below, previous field studies have found surprising results regarding particle size distributions of dust transported far from the source. Here, we applied our model to address whether these effects might be due to electrostatic forces on long-range dust transport.

Maring et al. (2003) compared the size distribution of aerosol samples collected in Izaña near the dust emission source with Puerto Rico after a transport time of 5.5 days from the source. In support, they modelled a 2000 m tall column of air typical of the Saharan Air Layer over the North Atlantic and Caribbean that has no vertical mixing and an initially uniform size distribution. When only taking into account gravitational and Stokes drag forces, their model predicted more large particles settled out than found in the transported aerosol samples. In contrast, their model showed agreement when they modified the terminal velocity with an arbitrary upward-contribution for all particles.

Using our model, we calculated the displacement of different-sized particles after transport for 5.5 days at terminal velocity to determine the fraction of particles removed due to settling using similar assumptions as Maring et al. (2003), but explicitly including electrostatic forces rather than an arbitrary upward velocity component. To simplify, we assumed that all the particles had the same polarity and magnitude of charge. As shown in Eq. 2, the electrostatic force on each particle depends on the particle surface charge density and ambient electric field strength. The actual surface charge density of lofted dust particles is unknown, but typical triboelectric charging values range over several orders of magnitude, from ~1 μC m^{-2} (Lee et al., 2018; Waitukaitis et al., 2015; Wang et al., 2019), to ~10 μC m^{-2} (Shen et al., 2016), and up to ~100 μC m^{-2} (Cottrell, 1978; Donald, 1968; Horn and Smith, 1992; Matsuyama et al., 2003; Miura and Arakawa, 2007; Nordhage and Bäckström, 1977). The maximum electrostatic charge on the surface of a material prior to gas breakdhaveown was found by Matsuyama (2018)) was found by Tatsuhi to be approximately 400 μC m^{-2} for a 10 μm particle. The magnitude of the electric field is also difficult to determine and can range over multiple orders of magnitude, with values ofup to 200 V m^{-1} in fair weather electric fields (Adlerman and Williams, 1996; Bennett and Harrison, 2007; Harrison, 2011), up to 15 to 2150 kV m^{-1} in dust storms (Bo and Zheng, 2013; Harrison et al., 2016; Jackson and Farrell, 2006; Schmidt et al., 1998; Zhang et al., 2018), and up to 500 kV m^{-1} during thunderstorms (Stolzenburg et al., 2007). Since it is difficult to determine both the charge density and the electric field can vary by orders of magnitude, we look at theconsider the product between the two, σE, to be the important parameter for determining the effects of the electrostatic forces. and the range of values possible given applicable values for charge density and electric field. Since both the surface charge density and the electric field strength can vary, we calculated the electrostatic forces based on the product of the surface charge density and the electric field, σE. Assuming an upper limit for surface charge density of 220 μC m^{-2}, which was found for triboelectrically charged quartz by Muira and Arakawa (2007), and a fair weather electric field strength up to 200 V m^{-1} (Harrison, 2011), the With thisGiven the range of possible electric fields and charge densities discussed above, the value of σE can range from 0.2 mC V m^{-3} to 200 C V m^{-3} up to 44 mC V m^{-3}; however,. It is far more likely for a particle to experienceparticles can only sustain a σE on the lower end of thise range where there are more consistent electric fields and easier to developmore easily developedprevalent charge densities. As shown in Fig. 5, when $\sigma E =$ 38 mC V m^{-3}, our model results are found to match well with the experimental data of Maring et al. (2003).

In order for electrostatics to facilitate long-distance transport, the charge on the particles and the electric field must subsist throughout transport. While charges on particles may decay through gas neutralization, experiments have found surfaces retaining charge even after days of exposure to ambient (Olthuis and Bergveld, 1992; Yuan and Li, 2005; Leonov et al., 2006) and charged dust has been observed in the atmosphere even after being transported for long times (Nicoll et al., 2011; Harrison et al., 2018; Renard et al., 2018a). Moreover, subsequent particle collisions during transport could further charge particles through the triboelectric effect. Electric fields strengths up to 200 V m^{-1} are naturally occurring in the atmosphere (Adlerman and Williams, 1996; Bennett and Harrison, 2007; Harrison, 2011) and can persist during dust transport. In addition, the presence of the dust particles can increase the electric field strength (Brazenor and Harrison, 2005; Ulanowski et al., 2007) and inclement weather can temporarily result in high electric field strengths (Harrison, 2011; Stolzenburg et al., 2007).Other field studies also found large particles to be enriched at higher elevations in comparison to models. Reid et al. (2003) used a particle measuring system mounted to an aircraft to measure dust size distributions in Puerto Rico and found that the ratio of large to small particles did not depend strongly on elevation. Furthermore, lidar studies of Saharan dust over Barbados (i.e., far from the source) suggested little variation in the particle size distribution between 1 and 4 km elevation, based on the near-constant value of the depolarization ratio and the similarity of the 355 nm, 534 nm, and 1085 nm laser results in this elevation range (Haarig et al., 2017).

We carried out simulations to investigate whether electric fields and the resulting electrostatic force on particles can lead to this enrichment of large particles at higher elevations. Particle diameters were based on a lognormal size distribution characteristic of atmospheric dust with $\mu_D = 1.5$ μm and $s_D = 0.75$ μm (Reid et al., 2003). ~~Particle surface charge densities~~The products of electric field and particle charge density were based on a normal distribution with $\mu_{\sigma E} = 0$ and $s_{\sigma E} =$ ~~110~~ 0, 11, and 22 ~~μ~~mC V m^{-}~~2~~3 such that half the particles~~ are of each polarity and highly charged particles (two standard deviations from the mean) have surface charge densities characteristic of triboelectrically charged quartz (Miura and Arakawa, 2007)~~experienced an electrostatic force in the upward direction, and half in a downward direction. ~~These values for $s_{\sigma E}$ are within the lower end of the range for σE described above; thus, we believe they are capable of being sustained through dust transport.~~ Simulations were carried out for 10^5 particles. As shown in Fig. 6, in the case of no ~~no electric field~~electrostatic forces, the average particle diameter is significantly larger at low elevations than at high elevations, in agreement with current models. In comparison, ~~reasonable fair weather electric field strengths can cause~~with $s_{\sigma E} = 22$ mC V m^{-3}, the average particle diameter ~~to~~ becomes ~~nearly~~ more constant with changing elevation.

Electrostatic forces act on charged particles such that those of one polarity are lifted to higher elevations while those of the opposite polarity fall to lower elevations. As a result, the elevation distribution of particles is stretched out and becomes more uniform. This leveling of the elevation distribution occurs for all sized particles, such that both large and small particles are more uniformly vertically distributed, causing the size distribution of particles to become more constant with changing elevation.

~~Moreover, Reid et al. (2003) found that the change in size distribution with elevation varied; on some days the size distribution would stay constant with elevation, on others it would vary such that large particles were lower and small particles were higher. The fair weather electric field strength is also prone to variation and depends on weather conditions (e.g. rain, fog, overcast etc.) (Harrison, 2011). Thus, electrostatic forces could help explain the variability of size/elevation distributions in addition to the minimal dependence of size distribution on elevation.~~

Several field studies have found larger particles transported than predicted when only taking into account gravitational and drag forces (Betzer et al., 1988; van der Does et al., 2018; Renard et al., 2018). Some studies have found particles with diameters greater than 40 μm and ranging to 450 μm transported long distances (van der Does et al., 2018; Renard et al., 2018b). In our modell~~l~~ed system, if electrostatic forces are neglected, any particle with a diameter greater than 7.3 μm would settle out of the 2000 m column within 5.5 days. However, from Fig. 7, which represents the ~~cumulative percentage~~size distribution of lofted particles~~ as a function of particle diameter~~, we see that electrostatic forces cause several particles larger than 7.3 μm to remain lofted, up to 17.6 μm. Even with the consideration of electrostatic forces, it is not expected that these much larger particles (40 to 450 μm) would be lofted.

Thus, it is likely that other forces contribute to the transport of large particles such as fast horizontal wind speeds, turbulence, and uplift in convective systems (van der Does et al., 2018).

For electrostatic forces to account for these deviations in size distributions of atmospheric dust particles, σE must be on the order of ~10 mC V m^{-3}. To the authors' knowledge, field studies of atmospheric dust have not measured surface charge densities of dust particles to confirm whether particles have sufficient charge for electrostatic effects to be significant; we note that volumetric charge densities have been measured (Nicoll et al., 2011), but cannot be translated to surface charge densities, which may be much larger than volumetric charge, which is the net charge of a distribution of negatively and positively charged particles~~ densities due to the cancelling effect of positively and negatively charged particles~~. However, we emphasize that surface charge densities on the order of ~10 to ~100 μC m^{-2}, which could cause significant electrostatic effects on transport, are routinely observed in laboratory settings (Cottrell, 1978; Donald, 1968; Horn and Smith, 1992; Matsuyama et al., 2003; Miura and Arakawa, 2007; Nordhage and Bäckström, 1977; Shen et al., 2016). While charges on particles may decay through gas neutralization, experiments have found surfaces retaining charge even after days of exposure to ambient conditions (Olthuis and Bergveld, 1992; Yuan and Li, 2005; Leonov et al., 2006) and charged dust has been observed in the atmosphere even after being transported for long times (Nicoll et al., 2011; Harrison et al., 2018; Renard et al., 2018a). Moreover, subsequent particle collisions during transport could further charge particles through the triboelectric effect.

Additionally, while electric fields up to 200 V m^{-1} are naturally occurring in the atmosphere (Adlerman and Williams, 1996; Bennett and Harrison, 2007; Harrison, 2011) and likely persist throughout transport, local electric fields within dust layers may be much higher due to the presence of dust particles and separation of positively and negatively charged particles (Brazenor and Harrison, 2005; Ulanowski et al., 2007). To the authors' knowledge, local electric fields within dust layers have not been measured over the course of long-distance dust transport. Thus, to better gauge the extent electrostatic forces influence dust transport, future field studies should investigate characteristic particle surface charge densities and local electric fields strengths within dust layers which can persist throughout long-distance dust transport.

6 Conclusion

We conducted experiments to characterize the size distribution as a function of height for sand blown between electrodes with vertically oriented electric fields and model this system considering the gravitational, electrostatic, and drag forces on particles. Our experimental and modeling results indicate that electrostatic forces maintain particles at higher elevations and increase the concentration of larger particles at higher elevations. We extend our model to long-distance dust transport and find that sufficient ~~that the presence of electric fields~~electrostatic forces~~ causes~~ suspend large particles that would otherwise settle out during transport, thereby increasing the concentration of large particles in atmospheric dust~~ particles to stay suspended that would otherwise settle out during transport. As large particles would preferentially settle out without electric fields, our results predict an increase in the concentration of lofted large particles~~. Since large particles have a heating effect due to absorption and scattering of radiation, our results suggest that electrostatic forces could contribute to the warming of the climate. In addition, we show ~~that dust transport models that incorporate electrostatic forces can better predict the~~that sufficient electrostatic forces may explain unexpected size~~/~~ and elevation distributions of atmospheric dust. ~~Thus, in order to accurately determine the effect of atmospheric dust on the climate, our results show electrostatic forces must be considered~~To better gauge the significance of electrostatic forces in atmospheric dust transport, future field studies should characterize particle surface charge densities and local electric fields subsisting in transported dust layers.

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Author Contributions

D. J. L., R. M. S., L. X., J. Z., and J. R. T. helped to design the experiments that J. R. T., S. R., H. S., and B. V. carried out. D. J. L. and S. R. designed the model and simulations that S. R. carried out. D. J. L., S. R., and J.R. T. analyzed and discussed the results. S. R., D. J. L., and J. R. T. wrote the manuscript. J. R. T., S. R., H. S., B. V., and D. J. L. contributed to the editing of the paper.

Competing Interests

The authors declare that they have no conflict of interest.

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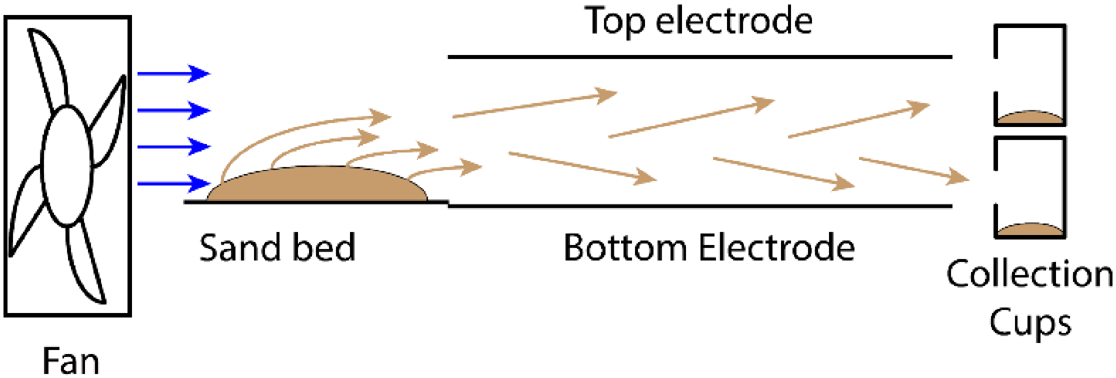

Figure 1: An experimental schematic of the dust system. A fan blows sand from a bed between two electrodes. Sand is collected in two collection cups after the electrodes.

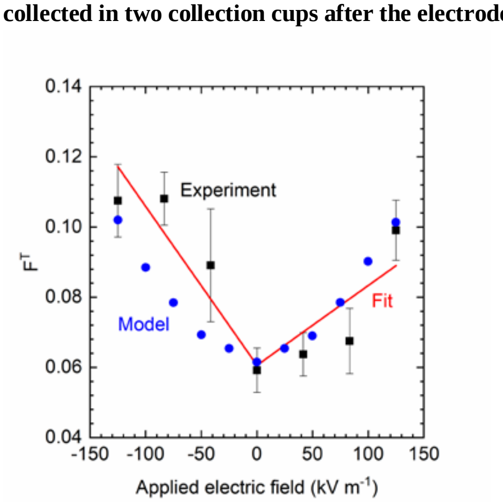

Figure 2: Plot of the fraction of total particles collected in the top cup (F^T) as a function of applied electric field. The black squares are the experimental results, the red line is a linear fit to the experimental data, and the blue circles are data from the model.

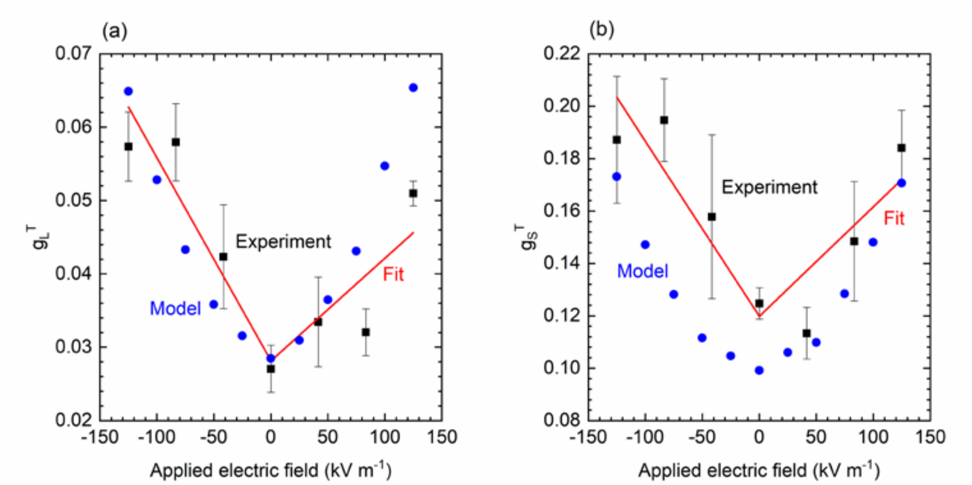

Figure 3: Fraction of total (top and bottom) large particles in the top cup (g_L^T) (a) and fraction of total (top and bottom) small particles in the top cup (g_S^T) (b) as a function of applied electric field. The black squares are the experimental results, the red line is a linear fit to the experimental data, and the blue circles are data from the model.

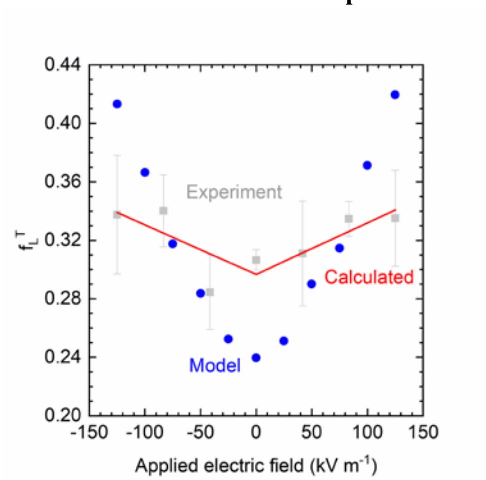

Figure 4: Fraction of large particles in the top cup (f_L^T). The black squares are the experimental results, the red line is the calculated values of f_L^T from Eq. (13), and the blue circles are data from the model.

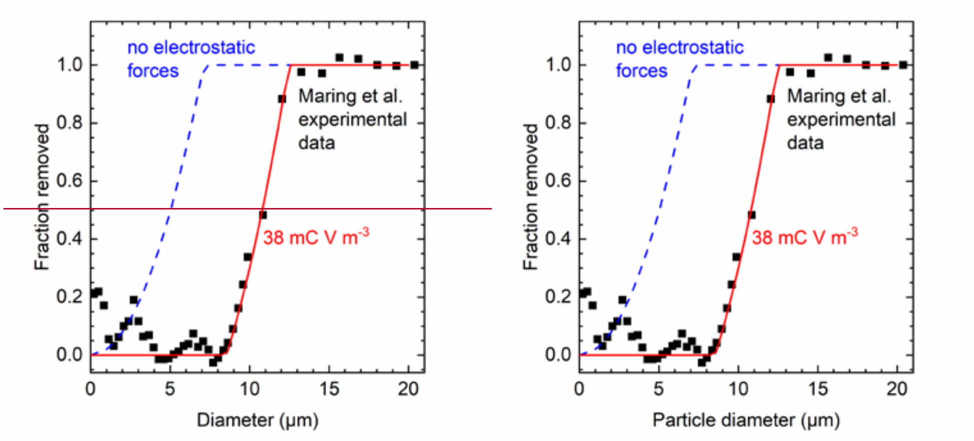

Figure 5: Fraction of particles removed by settling. The dashed blue line takes into account gravitational and drag forces, the red line takes into account gravitational, drag, and upward electrostatic forces, and the black points are experimental data collected from Maring et al. (200~~1~~3).

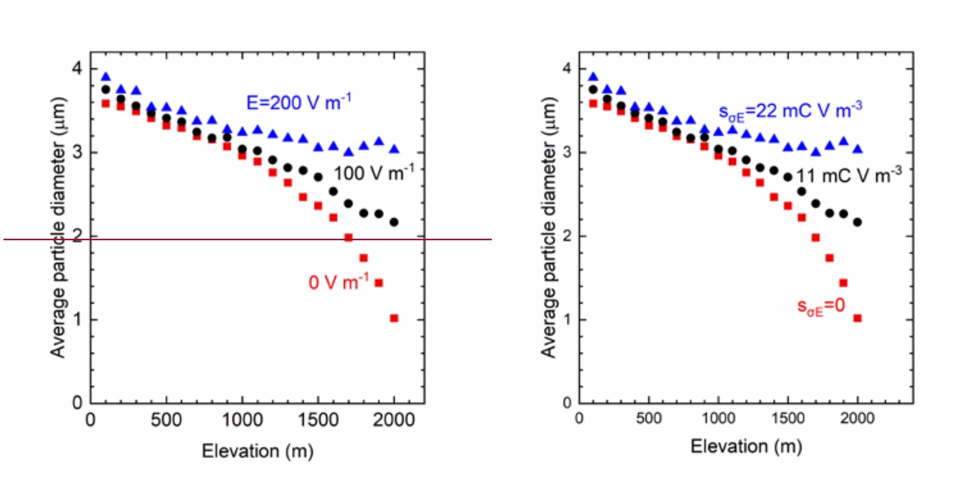

Figure 6: Model results for the average particle diameter as a function of elevation after 5.5 days of transport at various ~~electric field strengths~~standard deviations of electric field and particle charge density products, $s_{\sigma E}$.

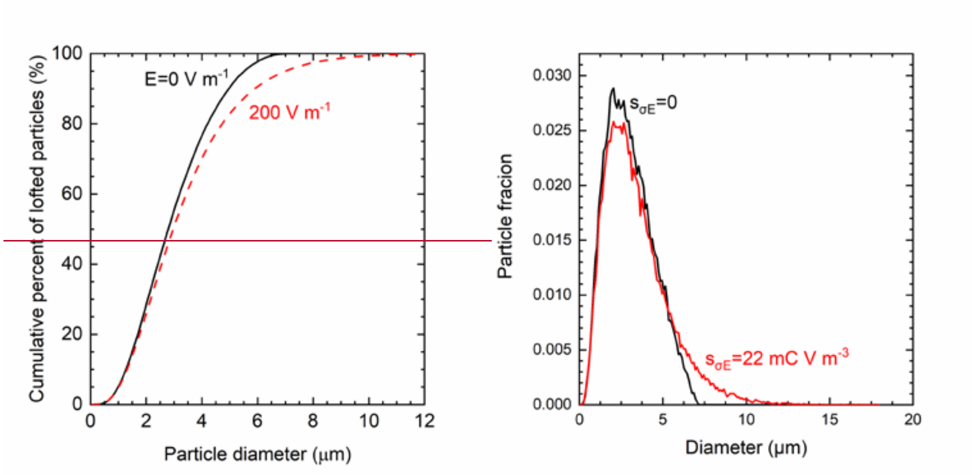

Figure 7: Remaining ~~Cumulative percent of lofted particles as a function of particle size at different electric field magnitudes~~particle size distributions within a 2000 m window after 5.5 days of transport at different values of $s_{\sigma E}$, with a bin size of 0.1 µm.~~.~~ The maximum size of lofted particles are 7.3 and 17.6 µm for ~~electric field strengths~~a $s_{\sigma E}$ of 0 and ~~200~~ 22 mC V m^{-1}, respectively.