Microphysical effects of relative humidity and temperature on the triboelectrification of volcanic ash

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Key Points:

• The triboelectrification of ash in low-energy collisions is modulated by humidity and temperature
• Both increasing temperature and humidity reduce the amount of charge collected by micron-sized particles
• The reduction in charging efficiency under humid environments could explain hiatuses in electrical activity at some volcanoes

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Abstract

Triboelectric charging has been cited as a principal electrification mechanism in volcanic granular flows. Previous work outside of the volcanic context has demonstrated that triboelectric processes are affected by environmental conditions. In particular, the amount of charge collected on surfaces in contact or undergoing frictional interactions appears to be controlled importantly by humidity and temperature. Here, we perform similar experiments using natural volcanic materials. By measuring the charge on individual micron-sized grains, we find that both increasing humidity and temperature decrease the efficiency of charge generation via triboelectric processes. Furthermore, we show that the effects of humidity on particle charging become important at relative humidities greater than 30%. Our work suggests that frictional electrification may be more efficient in the upper regions of the plume where the temperatures are low and the available water has been converted to ice.

1 Introduction

Volcanic plumes can become highly electrified as evidenced by changes in the ambient electric field beneath plumes [Nagata et al., 1946; Hatakeyama, 1947; Hatakeyama and Uchikawa, 1951; Lane and Gilbert, 1992; Miura et al., 1996], elevated charge densities detected on ash particles [Gilbert et al., 1991; Miura et al., 1996; Harrison et al., 2010], observations of tephra aggregates bound together by electrostatic forces [James et al., 2002, 2003; Telling et al., 2013], attenuation of L-band radiation propagating through plumes [Méndez Harper et al., 2019], and spectacular displays of lightning [Thomas et al., 2007; McNutt and Williams, 2010; Behnke et al., 2013; Behnke and Bruning, 2015; Aizawa et al., 2016; Cimarelli et al., 2016; Behnke et al., 2018; Méndez Harper et al., 2018a]. Field observations and experimental efforts suggest that electrostatic processes in plumes (a) arise from a wide range of charging and charge-separation mechanisms operating at different locations and times within the plume [James et al., 2000; Thomas et al., 2007; McNutt and Williams, 2010; Behnke et al., 2013; Behnke and Bruning, 2015; Méndez Harper et al., 2015; Méndez Harper and Dufek, 2016]; (b) reflect eruption kinematics [Behnke et al., 2013; Behnke and Bruning, 2015; Cimarelli et al., 2016; Méndez Harper et al., 2018a]; and (c) correlate with environmental conditions in the plume [Mather and Harrison, 2006; Arason et al., 2011; Nicora et al., 2013].
The mechanisms invoked to explain electrostatic processes in plumes include fragmentation charging [James et al., 2000; Méndez Harper et al., 2015], triboelectric charging [Houghton et al., 2013; Cimarelli et al., 2014; Méndez Harper and Dufek, 2016; Méndez Harper et al., 2017], charging arising from the decay of radioactive compounds in ash [Houghton et al., 2013], and mechanisms comparable to those thought to operate in conventional thunderclouds [Mather and Harrison, 2006; Nicora et al., 2013]. Triboelectrification—referring collectively to the electrification arising from frictional or collisional interactions between two or more surfaces—has been recognized since antiquity [Iversen and Lacks, 2012] and has been detected or inferred to exist in a number of natural granular systems in addition to volcanic plumes [Farrell et al., 2004; Fabian et al., 2001; Forward et al., 2009b; Harrison et al., 2016; Sternovsky et al., 2002; Forward et al., 2009d; Méndez Harper et al., 2017; Helling et al., 2013; Hodosán et al., 2016; Méndez Harper et al., 2018b]. Experimental studies have shown that volcanic ash readily charges triboelectrically [Hatakeyama and Uchikawa, 1951; James et al., 2000; Houghton et al., 2013; Cimarelli et al., 2014; Méndez Harper and Dufek, 2016; Méndez Harper et al., 2017]. Because pyroclast-pyroclast collisions occur within the plume throughout all stages of the eruption, triboelectrification likely contributes importantly to electrical processes in volcanic clouds.

Despite being one of the oldest manifestations of electricity known to humans, triboelectric charging involves underlying physical processes that remain inadequately understood [Lowell and Truscott, 1986; Lacks and Levandovsky, 2007; Lacks and Sankaran, 2011; Shinbrot, 2014]. For instance, experimental efforts have yet to unequivocally reveal the identities of the charge carriers being exchanged during collisions. Some investigators suggest that triboelectric charging arises from the exchange of electrons [Lowell and Rose-Innes, 1980; Lowell and Truscott, 1986; Lacks and Levandovsky, 2007; Forward et al., 2009a; Kok and Lacks, 2009], whereas other works point to an ionic process [Gu et al., 2013; Zhang et al., 2015; Xie et al., 2016]. Others still have argued that triboelectric charging requires the transfer of material from one surface to another (see, for example, Salanek et al. [1976]), implying that triboelectricity is nominally a manifestation of fragmentation charging.

Another aspect of triboelectric charging that remains poorly constrained involves the role of ambient conditions under which electrification takes place, specifically the presence of volatiles and temperature. While a number of investigators have attempted to elucidate how relative humidity (RH) influences triboelectric behavior, the results of these
studies sketch an inconclusive, sometimes contradictory, picture. Some studies report that charging decreases monotonically with increasing RH, citing an increase in air conductivity [Harper, 1957; Tada and Murata, 1995; Greason, 2000; Diaz and Felix-Navarro, 2004; Burgo et al., 2011; Schella et al., 2017]. Others report that low amounts of water actually enhance frictional electrification because H\(^+\) and OH\(^-\) tend segregate on large and small particles, respectively, producing the oft-cited size-dependent bi-polar charging [Hiratsuka and Hosotani, 2012; Gu et al., 2013; Xie et al., 2016]. In the volcanic context, James et al. [2000] reported that particles electrified via fragmentation and frictional processes were possibly influenced by relative humidity, but those authors indicate that their results may reflect decreases in the electrical impedance of their measurement equipment rather than changes in the amount of charge collected by the ash. While no consensus has been reached, a role of water in triboelectric charging is becoming increasingly difficult to dismiss.

Likewise, experimental work indicates that temperature has an effect on frictional charging processes. For example, Greason [2000] found that the magnitude of charge on beads in a vibrated bed decreases with increasing temperature. Additionally, Olsen et al. [2018] notes that a number of investigators (including Wen et al. [2014], Su et al. [2015], and Lu [2017]) report a power output decrease with temperature in triboelectric nanogenerators. Temperature variations may also affect the presence of contaminants on surfaces, changing their ability to retain charge [Lowell and Rose-Innes, 1980].

Grains in volcanic plumes are transported over a broad span of environmental conditions, possibly affecting their triboelectric characteristics. Erupted particles may have temperatures ranging from several hundreds of degrees near the vent to temperatures below 0\(^\circ\) C in the elevated, distal plume. Additionally, depending on the context of an eruption, ash particles may undergo collisions in relatively dry environments or be surrounded by humid air. However, the effects of these two environmental parameters on the electrification of plumes have been only minimally investigated experimentally (see James et al. [2000]). Here, we present the results of a set of experiments designed to elucidate the response of triboelectric charging of volcanic ash to changing relative humidity and temperature conditions. Specifically, we explore the conditions relevant to maturing plumes with temperature approaching the freezing point of water and low-energy particle-particle collisions. Our results show that triboelectric charging is highly sensitive to both humidity and temperature.
Table 1. Humidity and temperature conditions explored in the experiments

| Experiment   | Temperature | RH    |
|--------------|-------------|-------|
| Variable RH  | 20° C       | 0 – 50% |
| Variable T   | -20 – 40°   | 30%   |

2 Methods

To investigate the ability of ash particles to charge through frictional and collisional interactions given a variety of temperature/relative humidity regimes, we used the apparatus described in Méndez Harper et al. [2017] (shown schematically in figure 1). This apparatus allowed us to characterize the charge on single micron-sized ash grains. The device consists of a hollow aluminum tube open at one end and connected to the grounded shaft of a stepper motor at the other. The open end of the tube can be raised or lowered by a second stepper motor. At the onset of an experiment, the open end of the tube was raised 5 degrees from horizontal and 100 mL of particles were placed in its interior. The interior of the tumbler was coated with particles of identical size and composition such that tribocharging arises from particle-particle collisions only. To neutralize any charge that may have been transferred to the particles during handling, the sample in the tube was sprayed with a bipolar ionizing gun for 1 minute. The apparatus was then placed within a chamber in which we could control relative humidity and temperature. We performed two sets of experiments: 1) Varying humidity at a constant temperature and 2) varying temperature at a constant humidity. The environmental conditions we explored are summarized in table 1.

We allowed the conditions in the chamber to stabilize before starting an experiment. Once equilibrated, particles were charged by running the tumbler. Particles exchanged charge as they collided with each other and with the coated interior walls of the tube. The tube was spun at a rotation rate of 0.5 m/s for 20 minutes. This motion produced low-energy particle-particle collisions comparable to those found outside of the gas-thrust region. Likewise, the environmental conditions of the chamber are representative of conditions in elevated plume environments rather than those in the region proximal to the vent. Upon concluding the charging period, the rotating tube was inclined downward, causing the recently charged particles to roll out under gravity. Upon leaving the tube,
the particles passed through an open-ended Faraday cage connected to a charge amplifier similar to that described by Watanabe [2007]. To minimize problems associated with increased leakage currents caused by raising humidity (as reported in James et al. [2000]), we encased the Faraday cage in an acrylic conformal coating (see figure 1). The charge amplifier was maintained in a separate chamber to prevent drift in the electronics due to temperature changes.

The charge amplifier outputs voltage pulses, whose amplitudes, $V_o$, are proportional to the charges, $Q$, on individual particles falling through the sensing volume:

$$V_o = \frac{-Q}{C_f}.$$  

In equation 1, $C_f$ is the feedback capacitance of the charge amplifier, in this case 100 pF. The instrument can measure charges on individual particles as low as 1 fC (or $10^{-15}$ coulombs), enabling characterization of the magnitude and polarity of charge on individual ash particles. More details can be found in the supplemental document of Méndez Harper et al. [2017].

In these experiments, we employed ash from volcán Popocatépetl (“Smoking mountain” in Náhuatl, the lingua franca of pre-Colonial Mesoamérica), State of Puebla, México. The material was washed and sieved so as to have a nominal, spherical-equivalent particle size distribution between 125-250 µm. The material was washed with prior to running experiments to reduce the effects of contaminants and small fragments adhered to the surfaces of the particles.

3 Results

An example of raw data collected from the output of the charge amplifier during an experiment conducted at 20°C and 30% RH is provided in figure 2. Each pulse represents a charged particle passing through the Faraday cage. Very broad pulses or pulses with more than a single peak denote conditions in which more than one particle traversed the sensing volume at a time. We exclude such data from further analysis. In agreement with size-dependent bipolar charging (by which small particles gain negative charge with large particles acquire positive charge), most particles in our experiments acquired negative charge given that their surface areas are much smaller than the aggregate, interior
area of the coated tumbler [Forward et al., 2009b; Méndez Harper et al., 2017]. On average, grains obtained net charges on the order of 10^{-12} C for both sets of experiments.

A common way to quantify electrification in a granular substance is to compute the material’s surface charge density or the charge normalized by the particle’s surface area: \( \sigma = \frac{Q}{\pi D^2} \). For brevity, we use the nominal mean, spherical-equivalent diameter of the ash particles for all computations in this work (i.e. \( D \approx 188 \mu m \)). The mean charge densities observed in our experiments are on the order of 10^{-6} C m^{-2}. The relationships between temperature and relative humidity and charge density are rendered in figure 3a and figure 3b, respectively.

Our experiments show that both temperature and relative humidity have important effects on the magnitude of charge density acquired by ash particles in plumes. In agreement with other work, the degree of charging decreases monotonically with increasing relative humidity [Greason, 2000; Schella et al., 2017; Kolehmainen et al, 2027]. However, there appears to be a sharper decrease in the magnitude of charge density between 20-30 % RH. We also observe that charging of the volcanic sample decreases as the temperature increases. As discussed above, similar behaviors have been noted in non-volcanic materials by a number of investigators (e.g. Greason [2000], Su et al. [2015], Lu [2017], and Olsen et al. [2018]). Note that the effect of changing humidity is greater than that of changing temperature.

4 Discussion

Our data can be analyzed through the Greason equation, which describes the rate of triboelectric charging in a granular material [Greason, 2000]:

\[
\frac{dq(t)}{dt} = \alpha[q_s - q(t)] - \beta q(t) \tag{2}
\]

where \( q(t) \) is the charge on an ash particle at time \( t \), \( q_s \) is the maximum theoretical charge that can be sustained on a particle before the surrounding gas undergoes breakdown (for air, at 1 bar, the value of \( q_s = 2.66 \times 10^5 \pi D^2 \) coulombs is typically used), \( \alpha \) is a constant proportional to collision rate and the efficiency of charge exchange during a collision, and \( \beta \) encompasses charge loss mechanisms. Integrating the Greason equation, leads to:
\[ q(t) = q_0 e^{-(\alpha+\beta)t} + q_s \frac{1}{1 + \beta/\alpha} [1 - e^{-(\alpha+\beta)t}] \] (3)

Above, \( q_0 \) is any initial charge on grains prior to the triboelectric process (for instance, charge gained through fractoelectric charging). If \( t \to \infty \) and \( q_0 = 0 \), equation 3 reduces to:

\[ q_{ss} = \frac{2.66 \times 10^{-5} \pi D^2}{1 + \beta/\alpha} \] (4)

where \( q_{ss} \) is the steady-state charge. Alternatively, equation 4 can be expressed in terms of charge density by dividing the right-hand side by the particle’s surface area, \( \pi D^2 \):

\[ \sigma_{ss} = \frac{2.66 \times 10^{-5}}{1 + \beta/\alpha} \] (5)

Note that if \( \alpha \ll \beta \), \( \sigma_{ss} \to 0 \), while if \( \alpha \gg \beta \), \( \sigma_{ss} \to 2.66 \times 10^{-5} \text{ C m}^{-2} \), the breakdown limit for air at 1 bar.

Given our current understanding of triboelectric processes, however, it is difficult to determine whether the RH- and temperature-dependent behaviors we see in our experiments result from a reduced triboelectric charging efficiency (that is, a smaller \( \alpha \)), an increase in charge recombination processes (larger \( \beta \)), or both. One possibility is that humidity and temperature reduce the efficiency of charging (\( \alpha \)) by increasing the electrical conductivity of particles. Zheng et al. [2014] indicate that humidity control the stability of charge on particles through the deposition of water films containing dissolved salts. In other words, humidity modulates the surface conductivity of ash grains. Because natural silicate grains tend to have rough surfaces, adsorbed water molecules only form disconnected films that fill pores at low relative humidities. At some critical relative humidity, however, these films connect into a network across a particle’s surface, causing a precipitous increase in the surface conductivity. Previous work suggests that this transition occurs around 30-40% relative humidity. Zheng et al. [2014] propose that increased surface conductivity changes the energy barrier an electron must overcome in order to transfer from one surface to another during a particle-particle collision (this mechanisms, of course, only works if one assumes electrons are the principal species being exchanged during collisions). Those authors present a numerical model demonstrating that...
electron tunneling between surfaces is essentially cut off at the aforementioned critical
humidity. The steep drop in charge density observed in our experiments between 20 and
30% relative humidity hints that volcanic materials may follow this trend. Humidity may
also change the rate at which particles lose charge ($\beta$). While experimental data is sparse,
Tada and Murata [1995] and Méndez Harper et al [2019] provide evidence that humid
air may promote direct charge transfer to the atmosphere. However, future work needs
to be performed to elucidate the microphysical mechanisms underlying this process.

Unlike humidity, temperature may control both the surface and volumetric elec-
trical conductivities of materials. As mentioned above, temperature changes may mod-
ify the reactivity of contaminants on particle surfaces. However, because we carefully cleaned
the grains before conducting our experiments, we believe that changes in surface con-
ductivities were minimal. Thus, the decrease in charging efficiency with temperature could
indicate an increase in the bulk conductivity of the particles. Such trend has been ob-
erved in other silicate materials (especially those containing K, Ca, and Na) and has
been cited as evidence for an ionic conduction pathway in glasses [Eldin and El Alaily,
1998; Sasek and Meissnerová, 1981]. In essence, increasing temperature can reduce the
energy barriers that cations (positive ions) must overcome to migrate within the glass
network. The temperature-dependent volumetric conductivity $\rho$ (often expressed in units
of $\Omega \cdot m$ or $\Omega \cdot cm$) of oxide glasses has been described as:

$$\log \rho = A + B \frac{1}{T}, \quad (6)$$

where $A$ and $B$ are composition-dependent coefficients and $T$ is temperature in K.
While $A$ and $B$ are unknown for the ash employed here, we hypothesize that the temperature-
dependent conductivities of volcanic materials are qualitatively similar to those of well-
studied silicate glasses. This enhanced mobility of positive ions in volcanic ash could fa-
cilitate the recombination of electrons transferred during particle-particle collisions. Pres-
umably, such process would change how charge is partitioned and collected during sur-
face contacts (i.e. modulating the value of $\alpha$).

Given the uncertainties involved in determining $\alpha$ and $\beta$, we define the dimension-
less parameter $\gamma = \beta/\alpha$. The variation of $\gamma$ for both RH and temperature is rendered
in figure 4a and 4b, respectively. We find that the variation of $\gamma$ can be described by an
exponential of the form:
\[ \gamma = C_1 e^{c_2 x} + C_3 \] (7)

where \( x \) is either temperature or RH and \( C_1 \) through \( C_3 \) are constants. This equation is plotted as dotted curves in figures 4a and 4b. As can be seen, \( \gamma \) is always larger than 1 for the conditions employed in our experiments, indicating that charge-inhibiting mechanisms dominate over processes of charge accumulation.

5 The influence of humidity and temperature on the triboelectrification of ash in volcanic plumes

In volcanic ash clouds, both humidity and temperature change throughout the eruptive column. The reduction in the efficacy of triboelectric charging with both humidity and temperature may be congruent with the character of electrical storms observed at a number of volcanoes. Thomas et al. [2007], Behnke et al. [2013], and Behnke and Bruning [2015] report that the electrical phenomena during the Augustine (2006) and Redoubt (2008) eruptions (as measured with lightning mapping arrays) can be divided into two phases: 1) an explosive phase and 2) a plume phase. The first mode is cotemporaneous with forcing at the vent and involves the production of continual radio frequency emissions (CRF) in the gas-thrust region. The temperature range and particle collisional energies we explored in this work are not directly applicable to the near-vent environment. However, if the efficiency of triboelectric charging continues to decrease with temperature, the detection of strong CRF emissions at the vent may be representative of other electrification mechanisms such as fragmentation charging. The second mode of electrical activity observed at Augustine and Redoubt comprised large-scale (km-long) spark discharges in well-developed plumes. Electrification and discharge mechanisms similar to those observed in thunderstorms have been invoked to explain this form of volcanic lightning [Arason et al., 2011; Nicora et al., 2013; Hargie et al., 2018]. Between these two phases, Thomas et al. [2007], Behnke et al. [2013], and Behnke and Bruning [2015] note the existence of hiatuses in electrical activity (up to several minutes in length) as ash-laden flows transition from forced jets to buoyantly-rising plumes. The pause in electrical phenomena likely reflects a few processes. Firstly, charge generation through fragmentation charging becomes more difficult as disruptive and abrasive particle-particle collisions become less frequent and energetic with distance from the vent. Secondly, as delineated in Méndez Harper et al. [2018a], the rarefaction that occurs in the barrel shock
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of an expanding overpressured jet may lead to rapid charge loss from particle surfaces, resulting in a subsonic jet with lessened volumetric charge densities. Lastly, the experiments described here suggest that charge generation and/or retention may be compromised by the high relative humidity conditions that develop as a buoyantly-rising plume cools and entrains water from the environment. Under these high levels of relative humidity, the exchange of charge through contact electrification may be effectively inhibited despite the gains in charging efficiency offered by the drop in temperature with altitude [Greason, 2000; Zheng et al., 2014]. In other words, increasing humidity may have been partially responsible for the hiatuses in electrical activity observed at Augustine and Redoubt. The resumption of electrical activity in the mature plumes of the volcanoes likely indicates the freezing of magmatic and entrained water, catalyzing electrification mechanisms analogous to those found in thunderclouds. Additionally, the conversion of liquid water to ice could reinvigorate triboelectric charging as particles undergo ash-ice, ice-ice, and ash-ash collisions. Under cold temperatures aloft, our experiments indicate that such renewed frictional electrification could be more efficient than lower in the volcanic column.

6 Conclusions

In this work, we have described a set of experiments designed to explore the link between environmental conditions and the triboelectrification of volcanic ash on the scale of individual grains. Our work shows that the charge per unit area gained by ash particles during collisional and frictional interactions decreases with both increasing temperature and relative humidity. We found that, on average, humidity had a stronger effect than temperature on triboelectric charging. The hiatuses in electrical activity observed at a number of volcanoes (including Augustine and Redoubt) following the powerful CRF emissions that accompany explosive activity, may reflect, in part, the increasing humidity plumes as water condenses in the system. Renewed triboelectrification may occur at elevation in volcanic systems where the temperature is low and liquid water is converted to ice, helping to generate observed plume-phase electrical storms.

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Data availability statement: All data for this paper are contained in the main text of the present work (figures). Additional detailed description of experimental appara-
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References

Aizawa, K., Cimarelli, C., Alatorre-Ibarguengoitia, M., Yokoo, A., Dingwell, D., and Iguchi, M. (2016). Physical properties of volcanic lightning: Constraints from magnetotelluric and video observations at Sakurajima volcano, Japan. *Earth and Planetary Science Letters* 444:45–55.

Arason, P., Bennett, A. J., and Burgin, L. E. (2011). Charge mechanism of volcanic lightning revealed during the 2010 eruption of Eyjafjallajökull. *Journal of Geophysical Research: Solid Earth*. 116(B9).

Behnke, Sonja A. and Eric C. Bruning. (2015). Changes to the turbulent kinematics of a volcanic plume inferred from lightning data. *Geophysical Research Letters* 42(10):2015GL064199.

Behnke, Sonja A., Ronald J. Thomas, Stephen R. McNutt, David J. Schneider, Paul R. Krehbiel, William Rison and Harald E. Edens. (2013). Observations of volcanic lightning during the 2009 eruption of Redoubt Volcano. *Journal of Volcanology and Geothermal Research* 259:214–234.

Behnke, Sonja A., Harald E. Edens, Ronald J. Thomas, Cassandra M Smith, Stephen R. McNutt, A. R. Van Eaton, Corrado Cimarelli. (2018). Investigating the Origin of Continual Radio Frequency Impulses during Explosive Volcanic Eruptions. *Journal of Geophysical Research*

Burgo, T. A., Rezende, C. A., Bertazzo, S., Galembeck, A., and Galembeck, F. (2011). Electric potential decay on polyethylene: Role of atmospheric water on electric charge build-up and dissipation. *Journal of electrostatics*. 69(4), 401-409.

Cimarelli, C., M. A. Alatorre-Ibarguengoitia, K. Aizawa, A. Yokoo, A. Diaz-Marina, M. Iguchi and D. B. Dingwell. (2016). Multiparametric observation of volcanic lightning: Sakurajima Volcano, Japan. *Geophysical Research Letters* 43(9):2015GL067445.
Cimarelli, C., M. A. Alatorre-Ibarguengoitia, U. Kueppers, B. Scheu and D. B. Dingwell. (2014). Experimental generation of volcanic lightning. Geology 42(1):79–82.

Díaz, A. F. and Felix-Navarro, R. M. A semi-quantitative tribo-electric series for polymeric materials: the influence of chemical structure and properties. Journal of Electrostatics. 62(4) 277–290.

Eldin, F. and El Alaily, N. A. (1998). Electrical conductivity of some alkali silicate glasses Materials chemistry and physics. 52(2), 175–179.

Fabian, A., Krauss, C., Sickafoose, A., Horanyi, M., and Robertson, S. (2001). Measurements of electrical discharges in Martian regolith simulant. IEEE transactions on plasma science. 29(2): 288-291.

Farrell, W. M., Smith, P. H., Delory, G. T., Hillard, G. B., Marshall, J. R., Catling, D., ... and Cummer, S. A. (2004). Electric and magnetic signatures of dust devils from the 20002001 MATADOR desert tests. Journal of Geophysical Research: Planets. 109(E3).

Forward, Keith M., Daniel J. Lacks and R. Mohan Sankaran. (2009a). Charge Segregation Depends on Particle Size in Triboelectrically Charged Granular Materials. Physical Review Letters 102(2):028001.

Forward, K. M., Lacks, D. J., and Sankaran, R. M. (2009d). Triboelectric charging of lunar regolith simulant. Journal of Geophysical Research: Space Physics. 114(A10).

Forward, Keith M., Daniel J. Lacks and R. Mohan Sankaran. (2009b). Particle-size dependent bipolar charging of Martian regolith simulant. Geophysical Research Letters 36(13):L13201.

Gilbert, J. S., S. J. Lane, R. S. J. Sparks and T. Koyaguchi. (1991). Charge measurements on particle fallout from a volcanic plume. Nature 349(6310):598–600.

Greason, W. D. (2000). Investigation of a test methodology for triboelectrification. Journal of Electrostatics. (49) 0304-3886.

Gu, Zhaolin, W. Wei, J. Su and C. W. Yu (2013). The role of water content in tribo-electric charging of wind-blown sand Scientific reports 3:1337

Hargie, K., Van Eaton, A., Mastin, L. G., Holzworth, R. H., Ewert, J., and Pavolonis, M. (2018). Globally detected volcanic lightning and umbrella dynamics during the 2014 eruption of Kelud, Indonesia. Journal of Volcanology and Geothermal
Hatakeyama, H. (1947). On the Variation of the Atmospheric Potential Gradient caused by the Cloud of Smoke of the Volcano Asama. (The Fourth Report.). Journal of the Meteorological Society of Japan. Ser. II 25(1-3):39–39.

Hatakeyama, H. and K. Uchikawa. (1951). On the Disturbance of the Atmospheric Potential Gradient caused by the Eruption-smoke of the Volcano Aso. Papers in Meteorology and Geophysics 2(1):85–89.

Harper, W. R. (1957). The generation of static charge. Advances in Physics. 6(24), 365-417.

Harrison, R. G., Nicoll, K. A., Ulanowski, Z., and Mather, T. A. (2010). Self-charging of the Eyjafjallajökull volcanic ash plume. Environmental Research Letters 5(2): 024004.

Harrison, R. G., Barth, E., Esposito, F., Merrison, J., Montmessin, F., Aplin, K. L., ... and Houghton, I. M. (2016). Applications of electrified dust and dust devil electrodynamics to Martian atmospheric electricity. Space Science Reviews. 203(1-4), 299-345.

Helling, C., Jardine, M., Stark, C., and Diver, D. (2013). Ionization in atmospheres of brown dwarfs and extrasolar planets. III. Breakdown conditions for mineral clouds. The Astrophysical Journal. 767(2), 136.

Hiratsuka, K. I., and Hosotani, K. (2012). Effects of friction type and humidity on triboelectrification and triboluminescence among eight kinds of polymers. Tribology International. 55, 87-99.

Hodosán, G., Helling, C., Asensio-Torres, R., Vorgul, I., and Rimmer, P. B. (2016). Lightning climatology of exoplanets and brown dwarfs guided by solar system data. Monthly Notices of the Royal Astronomical Society. 461(4), 3927-3947.

Houghton, I. M., Aplin, K. L., and Nicoll, K. A. (2013). Triboelectric charging of volcanic ash from the 2011 Grímsvötn eruption. Physical review letters, 111(11), 118501.

Iversen, P., and Lacks, D. J. (2012). A life of its own: The tenuous connection between Thales of Miletus and the study of electrostatic charging. Journal of Electrostatics. 70(3), 309-311.

James, M. R., S. J. Lane and J. S. Gilbert. (2000). Volcanic plume electrification: Experimental investigation of a fracture-charging mechanism. Journal of Geophys-
James, M. R., J. S. Gilbert and S. J. Lane. (2002). Experimental investigation of volcanic particle aggregation in the absence of a liquid phase. *Journal of Geophysical Research: Solid Earth* 107(B9):2191.

James, M. R., Lane, S. J., and Gilbert, J. S. (2003). Density, construction, and drag coefficient of electrostatic volcanic ash aggregates. *Journal of Geophysical Research: Solid Earth* 108(B9).

Kok, J. F., and Lacks, D. J. (2009). Electrification of granular systems of identical insulators. *Physical Review E.* 79(5), 051304.

Kolehmainen, J., Sippola, P., Raitanen, O., Ozel, A., Boyce, C. M., Saarenrinne, P., and Sundaresan, S. (2017). Effect of humidity on triboelectric charging in a vertically vibrated granular bed: experiments and modeling. *Chemical Engineering Science.* 173, 363-373.

Lacks, Daniel J. and Artem Levandovsky. (2007). Effect of particle size distribution on the polarity of triboelectric charging in granular insulator systems. *Journal of Electrostatics* 65(2):107–112.

Lacks, Daniel J. and R. Mohan Sankaran. (2011). Contact electrification of insulating materials. *Journal of Physics D: Applied Physics* 44(45):453001.

Lane, S. J. and Gilbert, J. S. (1992). Electric potential gradient changes during explosive activity at Sakurajima volcano, Japan. *Bulletin of Volcanology.* 54(7), 590-594.

Lowell, J. and Rose-Innes, A. C. (1980). Contact electrification. *Advances in Physics.* 29(6), 947-1023.

Lowell, J and W. Truscott. (1986). Triboelectrification of identical insulators. I. An experimental investigation *Journal of physics D: Applied physics* 9(7):1273.

Lu, C. X., Han, C. B., Gu, G. Q., Chen, J., Yang, Z. W., Jiang, T., ... and Wang, Z. L. (2017). Temperature effect on performance of triboelectric nanogenerator. *Advanced Engineering Materials.* 19(12), 1700275.

Mather, T. A., and Harrison, R. G. (2006). Electrification of volcanic plumes. *Surveys in Geophysics.* 27(4), 387-432.

McNutt, Stephen R. and Earle R. Williams. (2010). Volcanic lightning: global observations and constraints on source mechanisms. *Bulletin of Volcanology* 72(10):1153–1167.
Méndez Harper, J. S., Dufek, J., and McAdams, J. (2015). The Electrification of Volcanic Particles during the Brittle Fragmentation of the Magma Column. *Proc. ESA Annual Meeting on Electrostatics 2015*.

Méndez Harper, J. and Dufek, J. (2016). The effects of dynamics on the tribo-electrification of volcanic ash. *Journal of Geophysical Research: Atmospheres* p. 2015JD024275.

Méndez Harper, J., Huang, T., and Burton, J. C. (2019). The longevity of electrostatic charge on suspended atmospheric dust grains *American Geophysical Union, Fall Meeting 2019*

Méndez Harper, J. S., McDonald, G. D., Dufek, J., Malaska, M., Burr, D., Hayes, A., McAdams, J., and Wray, J. (2017). Electrification of sand on Titan and its influence on sediment transport. *Nature Geoscience*. 10(4), 260.

Méndez Harper, J. S., Cimarelli, C., Dufek, J., Gaudin, D., and Thomas, R. J. (2018a). Inferring compressible fluid dynamics from vent discharges during volcanic eruptions. *Geophysical Research Letters*. 45(14), 7226-7235.

Méndez Harper, J. S., Helling, C., and Dufek, J. (2018b). Triboelectrification of KCl and ZnS particles in approximated exoplanet environments. *The Astrophysical Journal*, 867(2), 123.

Méndez Harper, J. S., Steffes, P., Dufek, J., and Akins, A. (2019). The effect of electrostatic charge on the propagation of GPS (L-band) signals through volcanic plumes. *Journal of Geophysical Research: Atmospheres*. 124 (4), 2260–2275.

Miura, Toshiro, Takehiro Koyaguchi and Yoshikazu Tanaka. (1996). Atmospheric electric potential gradient measurements of ash clouds generated by pyroclastic flows at Unzen Volcano, Japan. *Geophysical Research Letters* 23(14):1789–1792.

Nagata, T., Hirao, K., Fukushima, M., and Takahashi, T. (1946). The change in point-discharge current due to the volcanic smoke of Sakura-jima. *Bull. Earthq. Res. Inst.* 24, 221-227.

Nicora, M. G., Bürgesser, R. E., Rosales, A., Quel, E. J., and Ávila, E. E. (2013). Actividad eléctrica asociada a la erupción del complejo volcánico Cordón Caulle durante 2011. *Meteorológica*. 38(2), 121-131.

Olsen, M., Örtengren, J., Zhang, R., Reza, S., Andersson, H., and Olin, H. (2018). Schottky model for triboelectric temperature dependence. *Scientific reports*. 8(1), 5293.
Salaneck, W. R., Paton, A., and Clark, D. T. (1976). Double mass transfer during polymerpolymer contacts. *Journal of Applied Physics*, 47(1), 144-147.

Sasek, L. and Meissnerová, H. (1981). Electrical conductivity of silicate glasses and melts. *Silikaty*. 25(1), 21–34.

Schella, A., Herminghaus, S., and Schröter, M. (2017). Influence of humidity on tribo-electric charging and segregation in shaken granular media. *Soft matter*. 13(2), 394-401.

Shinbrot, T. (2014). Granular electrostatics: Progress and outstanding questions. *The European Physical Journal Special Topics*. 223(11), 2241-2252.

Sternovsky, Z., Robertson, S., Sickafoose, A., Colwell, J., and Horányi, M. (2002). Contact charging of lunar and Martian dust simulants. *Journal of Geophysical Research: Planets*, 107(E11), 15-1.

Su, Y., Chen, J., Wu, Z., and Jiang, Y. (2015). Low temperature dependence of triboelectric effect for energy harvesting and self-powered active sensing. *Applied Physics Letters*. 106(1), 013114.

Tada, Y., and Murata, Y. (1995). Direct charge leakage through humid air. *Japanese journal of applied physics.*, 34(4R), 1926.

Thomas, R. J., P. R. Krehbiel, W. Rison, H. E. Edens, G. D. Aulich, W. P. Winn, S. R. McNutt, G. Tytgat and E. Clark. (2007). Electrical Activity During the 2006 Mount St. Augustine Volcanic Eruptions. *Science* 315(5815):1097–1097.

Telling, J., Dufek, J., and Shaikh, A. (2013). Ash aggregation in explosive volcanic eruptions. *Geophysical Research Letters*. 40(10), 2355-2360.

Watanabe, H., Ghadiri, M., Matsuyama, T., Long Ding, Y., and Pitt, K. G. (2007). New instrument for tribocharge measurement due to single particle impacts. *Review of scientific instruments*. 78(2), 024706.

Wen, X., Su, Y., Yang, Y., Zhang, H., and Wang, Z. L. (2014). Applicability of triboelectric generator over a wide range of temperature. *Nano Energy*, 4, 150-156.

Xie, L. and Bao, N. and Jiang, Y. and Zhou, J. (2016) Effect of humidity on contact electrification due to collision between spherical particles. *AIP Advances* 6(3):2158-3226.

Zhang, Y., Pähtz, T., Liu, Y., Wang, X., Zhang, R., Shen, Y., ... and Cai, B. (2015). Electric field and humidity trigger contact electrification. *Physical Review X*. 5(1), 011002.
Zheng, X., Zhang, R., and Huang, H. Theoretical modeling of relative humidity on contact electrification of sand particles. *Scientific reports*, 4, 4399.
**Figure 1.** Schematic of laboratory apparatus, adapted from Méndez Harper et al. [2017]. Charge device consists of a hollow aluminum tube open at one end and connected to the grounded shaft of a stepper motor at the other. A. The open end of the tube is raised by a second stepper motor to enable particles to be input to the device. The interior of the tumbler is coated with particles of identical size and composition as the input particles such that tribocharging arises from particle-particle collisions only. B. The tube is shifted downward, still rotating, causing particles to fall out of the tube and into the Faraday cage where the charge on individual particles is measured.
Figure 2. Example of data collected from the charge amplifier as particles fall through the Faraday cage. Clear peaks and troughs represent the magnitudes of charge on individual particles. Misshapen peaks result from multiple particles passing through the Faraday cage are excluded from further analysis. Note that the majority of peaks are negative, resulting from the fact that free particles have areas which are smaller than the collective surface area of particles adhered to the tube. Experiment shown was conducted at 20 C and 30% RH.
Figure 3. A. Mean charge densities for experiments conducted at 25°C and relative humidity varying from 0 to 50%. B. Mean charge densities for experiments conducted at 30% RH and temperatures ranging from -20 to 40°C.
Figure 4. A. The variation of the dimensionless parameter $\gamma$ increases with increasing relative humidity. B. The variation of $\gamma$ increases with increasing temperature.